PREPARATION AND CHARACTERISATION OF FUNCTIONALLY GRADED PIEZOCERAMICS

Erling Ringgaard*, Tomasz Zawada*, Karsten Hansen*, Sidney B. Lang**
Guy Feuillard*** & Emmanuel Le Clézio***

*Ferroperm Piezoceramics A/S, Kvästgaard, Denmark, e-mail: erling.ringgaard@meggitt.com
**Department of Chemical Engineering, Ben-Gurion University of the Negev, Israel
***Laboratoire Imagerie et Cerveau, Université François Rabelais de Tours, FR CNRS 3110 U INSERM 930, France

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].

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.

In order to obtain a porosity gradient, Pz27 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 $T > 1240^\circ\mathrm{C}$ 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.

Experimetal work and microscopy

- 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 somewhat quantify the even harmonics, an empirical parameter $r$ 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: $r = \frac{\phi_i}{\phi_0} \left(\frac{d_i}{d_0}\right)^p$.
- In Table 1, $r$ and other characteristic parameters are summarised and compared to values obtained by thermal, directional depoling.

"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 ($r > 0.5$), 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.

Conclusions and outlook

- 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.

Electrical characterisation

- In the present work, two types of gradients 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.

Experimental work and microscopy

- 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 somewhat quantify the even harmonics, an empirical parameter $r$ 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: $r = \frac{\phi_i}{\phi_0} \left(\frac{d_i}{d_0}\right)^p$.
- In Table 1, $r$ and other characteristic parameters are summarised and compared to values obtained by thermal, directional depoling.

"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 ($r > 0.5$), 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.

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 ($r > 0.5$), 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


Acknowledgement

The support of the European Commission through the AISHA-II project (Contract No. AAT-2007-212912) is gratefully acknowledged.