Available online at www.sciencedirect.com

SciVerse ScienceDirect

Ceramics International 39 (2013) S587–S590

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

A simple thermal wave method for the evaluation of the polarization state of embedded piezoceramics

Gunnar Suchaneck[a,*], Agnes Eydam[a], Matthias Rübner[b], Matthias Schwankl[b], Gerald Gerlach[a]

[a]*Solid State Electronics Lab, TU Dresden–Helmholtzstrasse 18, 01062 Dresden, Germany*

[b]*Chair of Metals Science and Technology, Friedrich-Alexander-Universität Erlangen-Nuremberg-Martensstrasse 5, 91058 Erlangen, Germany*

Available online 23 October 2012

Abstract

In this work, we demonstrate that the thermal wave method is a promising approach for non-destructive evaluation of polarization state of embedded piezoelectrics in integrated sensor–actuator modules. The embedded piezoceramics are subjected to periodic heating by a square wave-modulated laser array through the top layers. A transient thermal analysis was performed using the finite element modeling package ANSYS. The ANSYS simulation reveals the presence of a transient heating-up period before reaching steady state. Inside the PZT piezoceramics, the thermal excitation becomes nearly sinusoidal and shifted in phase as known for harmonic excitation. At low modulation frequencies, the pyroelectric response of the embedded $Pb(Zr,Ti)O_3$ plates is governed by thermal losses to the embedding layers. Here, the sample behavior can be described by a harmonically heated piezoelectric plate exhibiting heat losses to the environment characterized by a single thermal relaxation time enabling an estimation of the thermal conductance at the interfaces of the embedded piezoelectric.

Keywords: B. Non-destructive evaluation; D. PZT; E. Actuators

1. Introduction

Transient methods for the determination of the thermal or pyroelectric properties of thin films by pulse or periodic heating the film have been known for more than 140 years [1–6]. Fourier's law and the law of energy conservation result in a parabolic heat diffusion equation which yields for a homogeneous and isotropic material in the absence of internal heat sources

$$\Delta\Theta - \frac{1}{\alpha}\frac{\partial\Theta}{\partial t} = 0 \qquad (1)$$

where Δ is the Laplace operator, $\Theta = T - T_0$, T_0 the temperature of the environment and α the thermal diffusivity.The solution of this equation for a slab with periodic surface temperature is a temperature oscillation which is attenuated and retarded in phase with increasing depth [1]. Such a temperature oscillation can be regarded as a highly damped wave exhibiting a complex wave vector [7]

$$k = \sqrt{\frac{i\omega}{\alpha}} = (1+i)\sqrt{\frac{\omega}{2\alpha}} \qquad (2)$$

which is determined by the circular frequency of heat modulation ω. Note, that following Fourier's law, the time average of the heat flux taken over a period is zero. Therefore, thermal waves are not traveling waves carrying energy, but just an oscillation of the temperature field [8]. According to eq. (2), these oscillations are damped already within one period (the "wavelength" of the temperature oscillations amounts to $\lambda = 2\pi \cdot d_p$, where $d_p = (2\alpha/\omega)^{1/2}$ is the penetration depth of the temperature oscillation). Consequently, by recording a heating-rate-dependent physical property such as the pyroelectric coefficient [4,5], the depth profile of this property may be determined from spectral measurements employing the frequency-dependent penetration depth d_p. This was demonstrated by the laser intensity modulation method (LIMM) which is suitable to

*Corresponding author. Tel.: +49 351 463 35281; fax: +49 351 463 32320.
E-mail address: Gunnar.Suchaneck@tu-dresden.de (G. Suchaneck).

0272-8842/$ - see front matter
http://dx.doi.org/10.1016/j.ceramint.2012.10.141

obtain polarization or space charge profiles in ferroelectrics and polymers [6].

The pyroelectric coefficient at constant stress is the sum of the pyroelectric coefficient at constant strain (primary effect) and the piezoelectric effect due to thermal strain (secondary effect) [9]

$$p_i^\sigma = p_i^\varepsilon + d_{ijk}^T c_{jklm}^T \delta_{lm}^\sigma \quad (3)$$

where $p_i^\sigma$and p_i^ε denote the pyroelectric coefficients measured at constant stress and strain, respectively; d_{ijk}^T represents the piezoelectric tensor at constant T, c_{jklm}^T the elastic stiffness at constant T and δ_{lm}^σ the thermal expansion tensor at constant σ. Eq. (3) is valid at constant electric field. The primary effect of PZT is opposite in sign to the much smaller secondary one [10]. In relaxor ferroelectric piezocrystals like PMN-PT, the piezoelectric effect due to thermal strain provides a significant contribution to pyroelectric response [11]. This provides the capability to control the poling procedure by means of periodic heating.

Recently, we have demonstrated that periodic heating by a modulated laser beam provides a promising approach for non-destructive evaluation of the polarization state in integrated sensor–actuator modules embedded into low temperature co-fired ceramics (LTCC) piezoelectrics [12,13], and in Macro-Fiber Composite (MFC) actuators consisting of piezoceramic rods embedded into epoxy resin at both sides, located between two copper electrode strips of the electrode structure and covered by a Kapton film [13]. With regard to eq. (2), the main difficulty for the evaluation of embedded piezoelectrics is the exponentially decaying amplitude of temperature oscillations in the embedding material making the pyroelectric signal weak and thus increasing measurement uncertainty. Therefore, up to 100 measurement repetitions should be used for averaging in order to reduce noise.

The low-frequency pyroelectric response (<10 Hz) is determined by thermal losses induced by heat conduction and radiation to the surroundings. It enables an estimation of the thermal relaxation time and thus of the thermal conductance at the interfaces of the embedded piezoelectric to the environment.

In this work, these results are generalized and extended to PZT plates embedded into casted aluminum.

2. Analytical and numerical modeling

The solution of the thermal problem of a slab with periodic surface temperature and constant temperature at the bottom surface is given by [8]

$$\Theta(z,t) = \Theta(z) + \Theta_\sim(z,t) \quad (4)$$

where $\Theta(z)$ is a time-independent temperature rise above ambient temperature and $\Theta_\sim(z,t)$ the periodic solution. For a periodically heated piezoelectric plate of thickness d_f showing a homogeneous pyroelectric coefficient p_0, the average charge induced due to the pyroelectric effect depends on the mean temperature and yields:

$$\langle Q \rangle = \langle p\Theta \rangle \approx \frac{p_0}{d_f} \mathrm{Re} \int_0^{d_f} \Theta(z) \exp(i\omega t) dz \quad (5)$$

In the case of square wave modulation with a period $T = 2\pi/\omega_0$, the periodic solution is given in form of an infinite series by [7,12]

$$\Theta_\sim(z) = \Theta_\infty \left[\frac{\tau_{th}}{1 + i\omega\tau_{th}} + 2\sum_{n=1}^{\infty} \cos\left(\frac{n\pi \times z}{d_f}\right) \frac{\tau_d/n^2}{1 + i\omega\tau_d/n^2} \right] \times \sum_{m=1}^{\infty} \frac{(-1)^{m-1}}{2m-1} \exp(im\omega_0 t) \quad (6)$$

with $\Theta_\infty = \Phi_0 / c\rho d_f$ the asymptotic value of the periodic solution at $t \to \infty$, Φ_0 the heat flux absorbed by the plate surface, $\tau_d = d_f^2/\pi^2\alpha$ the heat diffusion time, and τ_{th} the thermal relaxation time of the embedded PZT plate. Fig. 1 illustrates the charge meter (Kistler 5015A) output voltage of a PZT plate embedded between two LTCC layers at a modulation frequency of 30 Hz satisfying Eqs. (4) to (6).

By traveling through the top layer of the module packaging, higher harmonics of a square wave disappear and the thermal response gradually becomes sinusoidal [7,13]. This is demonstrated in Fig. 2 illustrating the modeled time evolution of Θ in the center of the different materials of a commercial M-8528-P2MFC sensor–actuator module (Smart Material Corp., Sarasota, USA) for a heat flux modulation frequency of 30 Hz. The transient thermal analysis of the MFC actuator was performed using the finite element modeling package ANSYS® 11.0.

Considering a piezoelectric plate of area A, the pyroelectric response is given by [6]:

$$I_\sim(\omega) = \frac{Ap_0}{d_f} \int_0^{d_f} \frac{\partial\Theta(\omega,z)}{\partial t} dz \quad (7)$$

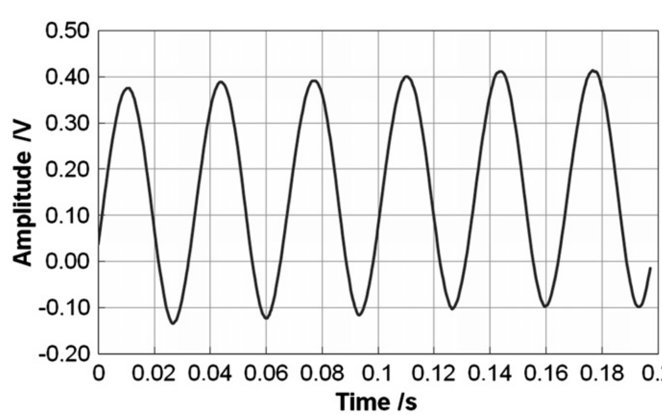

Fig. 1. Charge meter output voltage of an LTCC/PZT sensor–actuator module [13] at a modulation frequency of 30 Hz (DC-mode, time constant 10 s, low-pass filter 30 kHz).

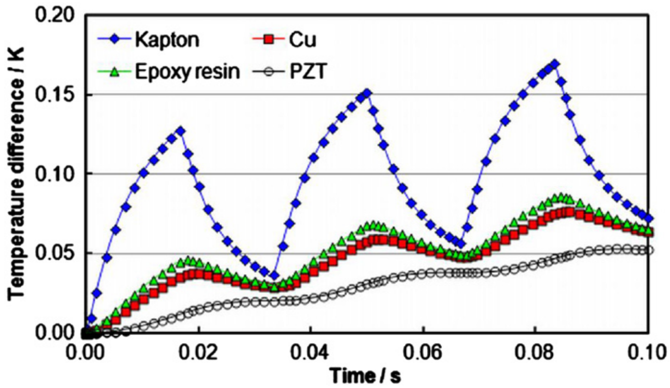

Fig. 2. Time evolution of the temperature difference to the environment in the center of the different MFC actuator materials for a heat flux modulation frequency of 30 Hz.

3. Experimental

For our experiments, two types of samples were evaluated:

(i). A LTCC/PZT module consisting of an already sintered PZT plate (CeramTecSonox® P53) with a size of $25\times10\times0.2\,mm^3$ embedded in the center of a $45\times20\times0.7\,mm^3$ sized sintered LTCC module (HeraeusHeraLock® Tape-HL2000). Sample fabrication is described in detail elsewhere [14]. The sample capacitance was 30 nF, the dielectric loss tangent amounted to about 2% at 10 kHz.

(ii). Integrated piezoceramic sensor–actuators manufactured by high pressure die casting. DuraAct™ piezoelectric patch transducers P876K001 of $39.5\times19\times0.5\,mm^3$ size consisting of a $15\times30\times0.2\,mm^3$ sized piezoceramic plate and a polyimide package (PI Ceramic GmbH, Lederhose, Germany) were fixed between sheets of expanded metal inside the mold onto two ejector pins. Casting was performed employing the casting alloy $AlSi_9Cu_3Fe$ (D226) at a mold temperature of 150 °C and a maximum during filling plunger velocity of 2 m/s. The casted plate had a dimension of $178\times178\times4\,mm^3$. The piezoelectric patch was located in the center at about 20 mm from the upper edge. Details of the fabrication process were reported in [15]. The piezoelectric patch capacitance was 25 nF, the dielectric loss tangent amounted to about 3% at 10 kHz. Samples with patch transducers located at the neutral axis as well as displaced off-axis were prepared.

The sensor–actuator samples were periodically heated by an array of 6 semiconductor lasers (LCU98A041A, Laser Components GmbH, Olching, Germany) square-wave-modulated with frequencies of up to 1 kHz each with a power of 14 mW at a wavelength of 980 nm. Polarization was determined from the pyroelectric current spectrum. The pyroelectric current was transformed into a voltage by a current-voltage-converter and amplitude and phase were determined by an impedance-phase analyzer (Solartron 1260, Solartron Analytical, Farnborough, UK). The current spectra were corrected by laser power measurements using internal photodiodes of the lasers. In order to reduce noise, up to 100 measurement repetitions were used for averaging.

4. Results and discussion

Fig. 3 compares the real and imaginary parts of the pyroelectric current of the LTCC/PZT and the die casted Al/polyimide/PZT modules with the dielectric patch located at the neutral axis in dependence on frequency for 50 measurement repetitions. Note, that in the latter case the signal is noisier because it was magnified by a factor of ten for illustration. In the LTCC/PZT case, the temperature oscillation is confined above 0.6 Hz within the sample. At frequencies above 10 Hz only the top LTCC layer is heated. Here, the pyroelectric response is described by a pyroelectric capacitor periodically heated by temperature oscillations from the LTCC top layer absorbed in the PZT plate top electrode. The pyroelectric response below 10 Hz is determined by thermal losses induced by heat conduction and radiation to the surroundings.

The maximum of the imaginary part of the pyroelectric current allows an estimation of the thermal relaxation time of the embedded PZT plate:

$$\tau_{th}=\frac{c\rho d_f}{2H}, \tag{8}$$

where H is the thermal conductance at the interface. Thermal relaxation time and the thermal conductance yield values of 0.135 s and 3650 W/m^{2}K, respectively.

In the Al/polyimide/PZT case, the real part of the pyroelectric current is of opposite sign compared to the LTCC/PZT case. Here, the signs of the real and imaginary

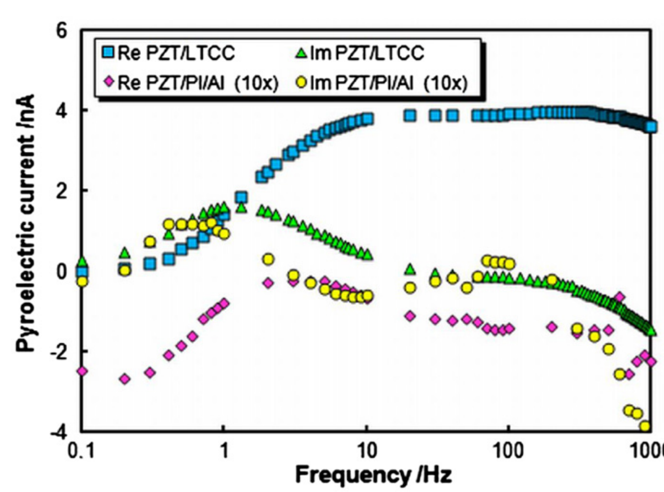

Fig. 3. Pyroelectric current spectrum of piezoelectric LTCC/PZT and die-casted Al/polyimide/PZT modules. For illustration, the values of die-casted Al/polyimide/PZT were magnified by a factor of ten.

parts of the pyroelectric current are sensitive to an off-axis location of the piezoelectric patch. Due to a larger heat capacity of the cast aluminum embedding, thermal losses occur already at by about one order of magnitude lower frequencies. On the other hand, the pyroelectric response decreases already at frequencies of less than 10 Hz because the penetration depth of temperature oscillations becomes smaller than the thickness of the embedding aluminum. At frequencies exceeding 170 Hz, the thermal diffusion time through the aluminum embedding becomes a limiting factor.

The maximum of the imaginary part of the pyroelectric current yields a thermal time constant of the embedded PZT plate of about 0.21 s in good agreement with a similar configuration of a metallized triglycine sulfate plate of a thickness of 0.1 mm on a 4 μm thick mylar sheet [16]. The corresponding thermal conductance amounts to about 2730 W/m^2K.

5. Conclusions

The thermal wave method was demonstrated to be a simple and non-destructive means for the evaluation of the polarization state of embedded piezoelectrics in integrated sensor–actuator modules. The determination of the thermal relaxation time of the embedded piezoelectrics enables disclosure of lamination failures.

Acknowledgments

This work was supported by the German Research Foundation (DFG)—Collaborative Research Center/ Transregio 39 "PT-PIESA", subproject C8.

References

[1] A.J. Ångström, New method of determining the thermal conductibility of bodies, Philosophical Magazine Series 4 (1863) 25–166 130–142.
[2] R.W. King, A method of measuring heat conductivities, Physical Review 6 (1915) 437–445.
[3] C. Starr, An improved method for the determination of thermal diffusivities, Review of Scientific Instruments 8 (1937) 61–64.
[4] A.G. Chynoweth, Dynamic method for measuring the pyroelectric effect with special reference to barium titanate, Journal Applied Physics 27 (1956) 78–84.
[5] H.J. Zajosz, Pyroelectric response to step radiation signals in thin ferroelectric films on a substrate, Thin Solid Films 62 (1979) 229–236.
[6] S.B. Lang, D.K. Das-Gupta, Laser-intensity modulation method: a technique for determination of polarisation and space charge in polymer electrets, Journal Applied Physics 59 (1986) 2151–2160.
[7] H.S. Carslaw, J.C. Jaeger, Conduction of Heat in Solids, 2nd ed, Oxford Univ. Press, New York, 1959.
[8] A. Salazar, Energy propagation of thermal waves, European Journal Physics 27 (2006) 1349–1355.
[9] J.F. Nye, Physical properties of crystals: their representation by tensors and matrices, Clarendon Press, Oxford, 1960.
[10] W.R. Cook, D.A. Berlincourt, F.J. Scholz, Thermal expansion and pyroelectricity in lead titanate zirconate and barium titanate, Journal Applied Physics 34 (1963) 1392–1398.
[11] A. Movchikova, G. Suchaneck, O.V. Malyshkina, B.B. Pedko, G. Gerlach, Thermal wave study of piezoelectric coefficient distribution in PMN-PT single crystals, Advances in Applied Ceramics 109 (2010) 131–134.
[12] G. Suchaneck, W. Hu, G. Gerlach, M. Flössel, S. Gebhardt, A. Schönecker, Nondestructive evaluation of polarization in LTCC/PZT piezoelectric modules by thermal wave methods, Ferroelectrics 420 (2011) 25–29.
[13] G. Suchaneck, W. Hu, G. Gerlach, Evaluation of polarization of embedded piezoelectrics by the thermal square wave method, in: Proceedings of 20th IEEE International Symposium on Applications of Ferroelectrics and International Symposium on Piezoresponse Force Microscopy and Nanoscale Phenomena in Polar Materials, ISAF-2011-PFM Joint Conference, July 24–27, 2011, Vancouver, Canada, doi: 10.1109/ISAF.2011.6014120.
[14] M. Flössel, S. Gebhardt, A. Schönecker, A. Michaelis, Development of a novel sensor–actuator-module with ceramic multilayer technology, Journal of Ceramic Science Technology 1 (2010) 55–58.
[15] M. Rübner, C. Körner, R.F. Singer, Integration of piezoceramic modules into die castings—procedure and functionalities, Advances in Science and Technology 56 (2008) 170–175.
[16] A. Hadni, R. Thomas, Laser study of reversible nucleation sites in triglycine sulphate and applications to pyroelectric detectors, Ferroelectrics 4 (1972) 39–49.