Fenu, N.G., Giles-Donovan, N. and Cochran, S. (2020) BNT and Mn:PIN-PMN-PT Single-Sample Characterisation at Operational Temperature Range for High-Power Ultrasonic Applications. 2020 Joint Conference of the IEEE International Frequency Control Symposium and International Symposium on Applications of Ferroelectrics (IFCS-ISAF), 19-23 Jul 2020.

BNT and Mn:PIN-PMN-PT single-sample characterisation at operational temperature range for high-power ultrasonic applications

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Abstract— **Rapid and convenient sample characterization is an important capability in new material development and provides crucial data for activity such as device design. Lack of such capabilities can lead to unsophisticated proxies for overall material performance, such as the use of d_{33} to indicate material quality. This is a particular problem in piezoelectric materials, where the gold standard characterization technique requires multiple samples. In the present work, in contrast, BNT lead-free piezoceramic and Mn:PIN-PMN-PT piezocrystal have been fully characterized with a single-sample technique combining impedance spectroscopy with FEA simulation. The same technique has been adopted to obtain the full elastic-piezoelectric-dielectric matrix at temperatures up to 80°C. This technique has proven to be a valid alternative for transducer designers to obtain the necessary properties to run FEA simulations on new materials with an accurate material dataset, and predict device behaviour at high temperature.**

Keywords*— PIC 700, Lead-Free, BNT, Single-Crystals, Mn:PIN-PMN-PT, Material Characterisation, Laser Doppler Vaibrometry, Optimisation, RUS, Single-Sample, Ultrasound Material.

I. INTRODUCTION

Lead-based piezoelectric single crystals are emerging as an alternative to conventional PZT ceramic for applications of high power ultrasound. Among them, third generation single crystals such as the doped ternary compound Mn:PIN-PMN-PT exhibit $d_{33} > 1100$pC/N, $k_{33} > 0.9$, and $Q_m > 700$. These contribute to extremely large figures of merit (i.e k^2Q, dQ) when compared to piezoelectrically hard PZT [1, 2, 3].

In addition, the recent European Regulation of Hazardous Substances (RoHS and RoHS2), is increasingly regulating the presence of lead in electronic components [4]. This makes lead-free piezoceramic an important future alternative, with some compositions already commercially available, such as BNT – PIC 700 (PI Ceramic, Lederhose, Germany).

In ultrasonic transducer design, finite element analysis (FEA) has become the standard tool to design, predict and optimise device performance by enabling the designer to explore virtual prototypes in an inexpensive and quick way. FEA simulations require a complete set of material properties to run. Specifically, for piezoelectric material the full elastic-piezoelectric-dielectric (EPD) matrix is required as input.

Accurate material properties are crucial in FEA to obtain accurate simulation results. Moreover, transducer designers rely on few documents reporting piezomaterial datasets to build up the FEA input file. These datasets are common for PZT materials, which have been available for at least several decades, but obtaining them for piezocrystals, or even newer lead-free compositions, is often a challenge. Performing full and accurate characterisation of a piezoelectric material requires the purchase of an expensive sample set, laboratory resources and skills that a transducer designer does not have.

For piezoceramic with 6mm symmetry, the EPD matrix is constructed with 10 independent coefficients, which are obtained in the standard way by impedance spectroscopy requiring four different sample geometries [5, 6]. For piezocrystals with 4mm symmetry, the EPD matrix contains 11 independent constants with five samples required for the measurements. Because of material inconsistency, the standard multisample characterisation can lead to mismatch between FEA simulated and physical devices.

In applications requiring high power, piezoelectric materials often experience thermal stresses, which may affect material properties and behaviour [7, 8]. Therefore, to characterise piezomaterials under different temperature is particularly relevant.

In this paper, a single-sample characterisation method is presented as an alternative to the standard multisample method. This method can also be used to determine the EPD matrix at different temperatures, thus enabling the possibility to simulate device performance under different conditions.

II. METHODS AND RESULTS

A sensitivity analysis has been used to show that the resonance modes are sensitive to the EPD matrix coefficients in a sample with dimensions 7.5 x 7.5 x 7.5 mm^3 and a frequency range selected for parameter optimization. Fig. 1 shows the sensitivity of elastic constants c^E_{11}, c^E_{12}, and c^E_{13}, in Mn:PIN-PMN-PT as resonance and antiresonance peaks in the electrical impedance curve. Small alterations of these coefficients have a significant effect on resonant and antiresonant frequencies. Subsequently, elastic constants, permittivity and electrical impedance of both PIC 700 and Mn:PIN-PMN-PT have been determined to provide a first estimate for the optimization algorithm.

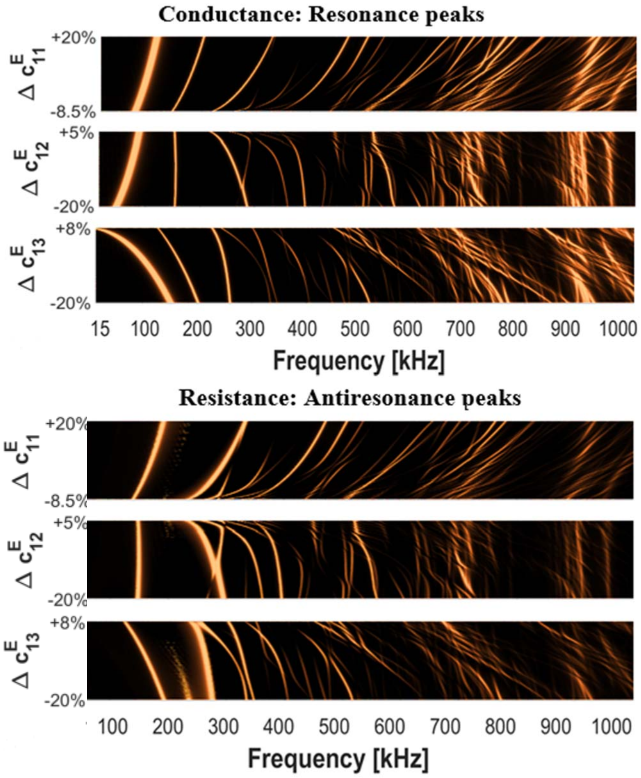

Figure 1: Sensitivity analysis of c^E_{11}, c^E_{12}, and c^E_{13} in Mn:PIN-PMN-PT cube.

In this study the Levenberg–Marquardt (LM) and Nelder–Mead (NM) optimization approaches were explored, with measured and simulated electrical impedance compared and matched in order to obtain coefficients in a fully automated MATLAB script combined with PZFlex FEA (OnScale, Cupertino, CA, USA). Validation of the refined parameters was performed by comparing the simulated modes with those reconstructed by adopting laser Doppler vibrometry on the sample's surface and measuring displacement and phase. Figure 2 shows an example of the results in the form of a vibration mode of the Mn:PIN-PMN-PT cube sample.

Figure 3 reports the results of the EPD matrix coefficients temperature characterisation from 20 to 80 degrees for PIC 700. After measuring the impedance of the sample at different temperatures, these curves have been fed into the optimization algorithm to obtain a full set of properties.

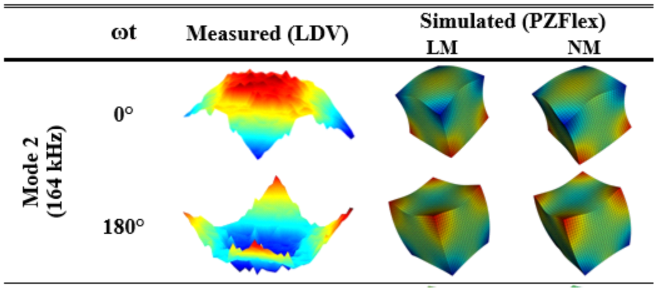

Figure 2: Mode reconstruction in Mn:PIN-PMN-PT cube.

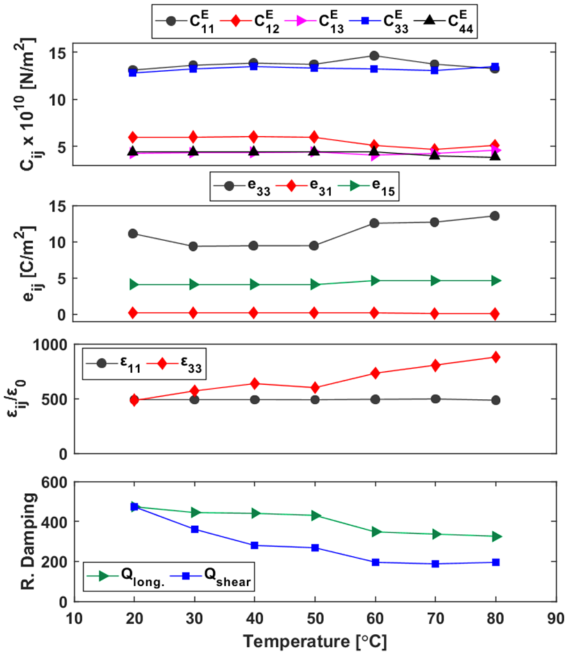

Figure 3: PIC 700 EPD coefficients at different temperatures obtained through optimization algorithm.

III. CONCLUSIONS

We have outlined a technique that requires only one piezoelectric sample to provide data allowing end-user oriented FEA modelling of piezoelectric materials and have demonstrated it on relaxor-type single crystals and lead-free material at room temperature and at elevated temperatures that may be found in power ultrasonics applications. We expect this to expedite translation from new materials research to future uses.

REFERENCES

[1] S. Zhang et al. "Advantages and challenges of relaxor-PbTiO3 ferroelectric crystals for electroacoustic transducers – A review" *Prog Mat Sci* 68, pp. 1 – 66, 2015 https://doi.org/10.1016/j.pmatsci.2014.10.002

[2] E. Sun & W. Cao "Relaxor-based ferroelectric single crystals: Growth, domain engineering, characterization and applications" *Prog Mat Sci* 65, pp. 124 – 210, 2014 https://doi.org/10.1016/j.pmatsci.2014.03.006

[3] S. Zhang and F. Li "High performance ferroelectric relaxor-PbTiO3 single crystals: Status and perspective" *J Appl Phys* 111 (3), 031301, 2012 https://doi.org/10.1063/1.3679521

[4] A. Mathieson and D. Deangelis, "Feasibility of lead-free piezoceramic based power ultrasonic transducers," *IEEE International Ultrasonics Symposium, IUS*, 2014

[5] T. Kinsley *et al.*, "IRE Standards on Piezoelectric Crystals : Measurements of Piezoelectric Ceramics , 1961 *," 1961.

[6] L. Pardo, 5 - Piezoelectric ceramic materials for power ultrasonic transducers, vol. 1920. Elsevier Ltd., 2015.

[7] X. Liao *et al.*, "Functional piezocrystal characterisation under varying conditions," *Materials (Basel).*, vol. 8, no. 12, pp. 8304–8326, 2015.

[8] X. Liao, T. Jiang, Z. Huang, and S. Cochran, "Loss characterisation of piezocrystals under elevated environmental conditions," *2016 Jt. IEEE Int. Symp. Appl. Ferroelectr. Eur. Conf. Appl. Polar Dielectr.*