Supplementary Information

for

Effect of inactive volume on thermocouple measurements of electrocaloric temperature change in multilayer capacitors of 0.9Pb(Mg_{1/3}Nb_{2/3})O₃-0.1PbTiO₃

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MLC fabrication

0.9PMN-0.1PT MLC samples were prepared by the conventional doctor blade method¹⁷. In order to reduce the parasitic pyrochlore phase, 0.9PMN-0.1PT powder was synthesized by the columbite method. The MgNb₂O₆ precursor was prepared from MgCO₃ (Mg 5 mol% excess) and Nb₂O₅. The starting powders were ball milled in distilled water with partially stabilized zirconia (PSZ) balls for 16 hours. After calcination at 1200 °C for 12 hours, the MgNb₂O₆ obtained was mixed with Pb₃O₄ (Pb 2 mol% excess), TiO₂ and PSZ balls, and milled for 24 hours. To obtain 0.9PMN-0.1PT powder, the slurry was dried and calcined at 750 °C for 4 hours. The 0.9PMN-0.1PT powder was then ball milled in an organic solvent with binder for 24 hours, and green sheets of 0.9PMN-0.1PT were fabricated by the doctor blade method with a 300 µm gap. After screen printing Pt paste for the inner electrode, green chips of 0.9PMN-0.1PT MLC were obtained by stacking, pressing and cutting the green sheets. The binder was burned at 500 °C for 4 hours, and the green chips were sintered at 1200 °C for 4 hours in a Pb-containing atmosphere. External electrodes were fabricated by silver paste.

MLC characterization

Using X-ray diffraction (RIGAKU, Rint-2000), the volume fraction of the parasitic pyrochlore phase was estimated from the intensities of the 110 perovskite reflection and the 222 pyrochlore reflection. A cross-sectional optical microscopy image [Fig. 1(b)] demonstrates that the inner electrodes are continuous, but this is not quite apparent from scanning electron microscopy (ELIONIX, ERA-8900) [Fig. S1(c)], due to the damage done when the MLC was broken to expose its interior (grains cannot be seen clearly after polishing).



Fig. S1. MLC structure. (a) Schematic and (b,c) cross-sectional images obtained by (b) optical microscopy and (c) scanning electron microscopy.

Measurement setup

The thermocouple was attached to the MLC face with kapton tape as shown below. These items were absent when measuring with the IR camera.



Fig. S2. Measurement setup with the thermocouple. (a) Photograph. (b) Schematic.

Dependence of measured temperature change on MLC geometry



Fig. S3. Dependence of measured temperature change on MLC geometry. Variation of $|\Delta T_j|$ with (a) active volume and (b) inactive volume, for an active area per layer of 0.12, 0.29 and 0.42 cm².