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Supplementary information

Broadening Operating Temperature Span of Electrocaloric Effect in Lead-Free Ceramics via Constructing Multi-stage Phase Transitions

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Sample characterization in detail

The schematic diagram of the EC test system is shown in Fig. S1a, where the specific parameters of the materials involved are given in Table S1. During the test, there is a heat exchange between the ceramic and the environment along the silver wire which results in heat loss. In order to obtain the intrinsic temperature change of the material, it is necessary to calculate the calibration factor (K) which can be described by:

$$K = \frac{\Delta T_{sp}}{\Delta T_{pt}} \tag{1}$$

where T_{sp} and T_{pt} denote the actual temperature change value of the sample and the temperature change value collected by PT100, respectively. A 1:1 test system model was built in COMSOL to realize this process and the results are shown in Fig. S1b. Taking L3 as an example, an initial EC temperature change of 1.28 K was assigned to the sample, while the PT100 detected a temperature drop of 0.68 K. The simulated temperature curve is in good agreement with the measured curve. The calibration factor in this work is 1.88.

Fig. S1. (a) Schematic diagram of the EC test system. (b) The temperature-time curves of the samples and PT100 calculated by COMSOL, where the measured curves are given for comparison.

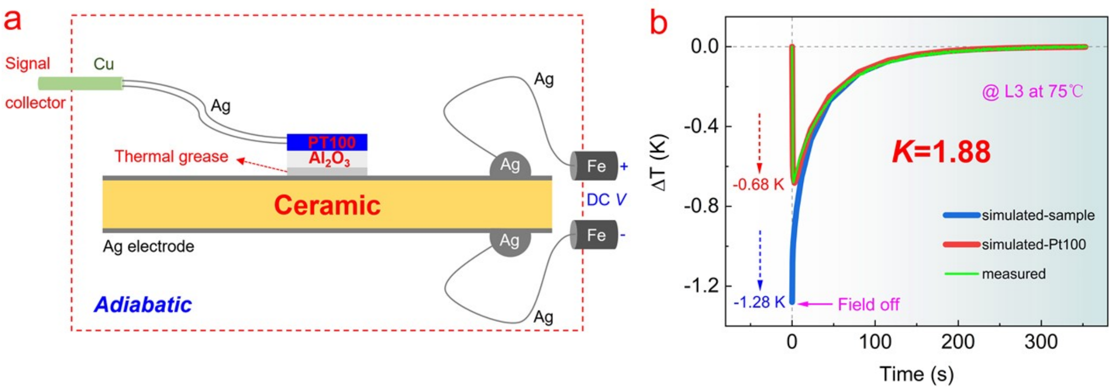

Table S1. Parameters of the materials involved in the ECE test system, where ρ, λ and *C*p represent density, coefficient of thermal conductivity and heat capacity at constant pressure, respectively.

Material	**ρ (kg m^{-3})**	**λ (W m^{-1} K^{-1})**	***Cp* (J kg^{-1} K^{-1})**	**Size**
Ceramic	5500	1.5	499	4.5 mm × 4.5 mm × 0.39 mm
Silver	10490	429	232	4.5 mm × 4.5 mm × 0.02 mm (electrode) Φ0.15 mm × 50 mm (connecting electrode) Φ0.15 mm × 10 mm (connecting PT100)
Thermal grease	2600	2	1083	1.7 mm × 1.2 mm × 0.2 mm
Aluminum oxide	3500	35	600	1.7 mm × 1.2 mm × 0.4 mm
Pt100	21450	71	140	1.7 mm × 1.2 mm × 0.4 mm
Copper	8900	401	390	Φ0.6 mm × 100 mm
Iron	7860	80	460	Φ3 mm × 7.5 mm

Fig. S2. Change rate of temperature dependent EC response for single component ceramics and composite ceramics.

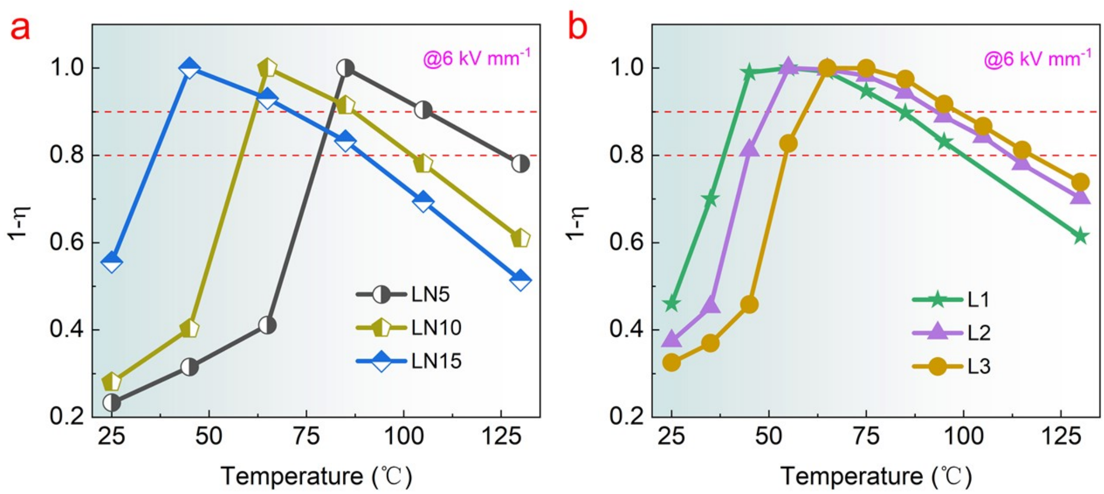

Fig. S3. Temperature dependence of P_m, P_r and P_m-P_r under an electric field of 6 kV mm^{-1}.