SUPPLEMENTARY MATERIAL

Tailoring the microstructure by a proper electric current control in flash sintering: the case of barium titanate

Samuel López-Blanco,[1] Diego A. Ochoa,[1] Xavier Vendrell,[2] Lourdes Mestres,[2] and Jose E. García[1]

[1] *Department of Physics, Universitat Politècnica de Catalunya – BarcelonaTech, 08034 Barcelona, Spain.*

[2] *Department of Inorganic and Organic Chemistry, Universitat de Barcelona, 08028 Barcelona, Spain.*

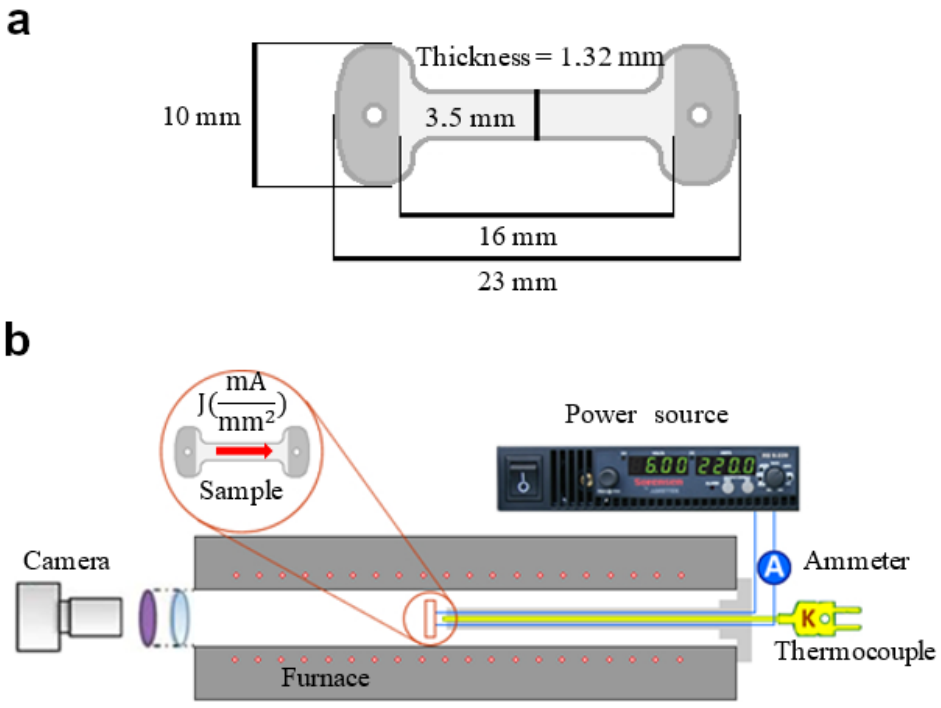

Fig. S1. (a) Green sample dimensions after uniaxial pressing and (b) typical flash sintering experimental setup scheme. Dark grey regions in the sample represent platinum painted electrodes.

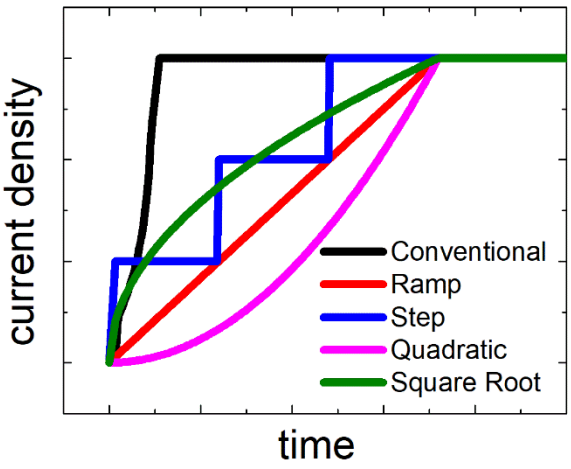

Fig. S2: Theoretically proposed current density profiles. Experiments are performed at a given temperature that depends on the electric field employed. Profile slopes are selected so that every experiment reaches maximum current density in the same time (except the conventional flash experiment, in which the sample undergoes current runaway almost instantly). Samples are then left to dwell for a set time.

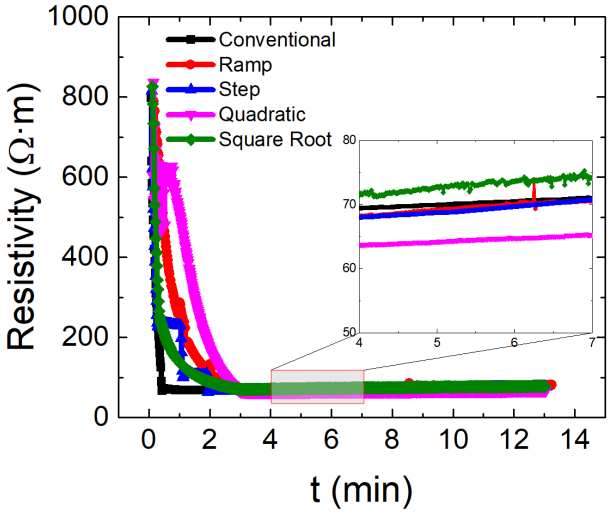

Fig. S3. Resistivity evolution of samples sintering with different electric current profiles, at 150 V/cm and 15 mA/mm^2. As shown, the initial values of the samples resistivity are quite different because different current profiles are used. During the dwell time, however, when samples have the same electrical conditions, all samples show similar values of resistivity.

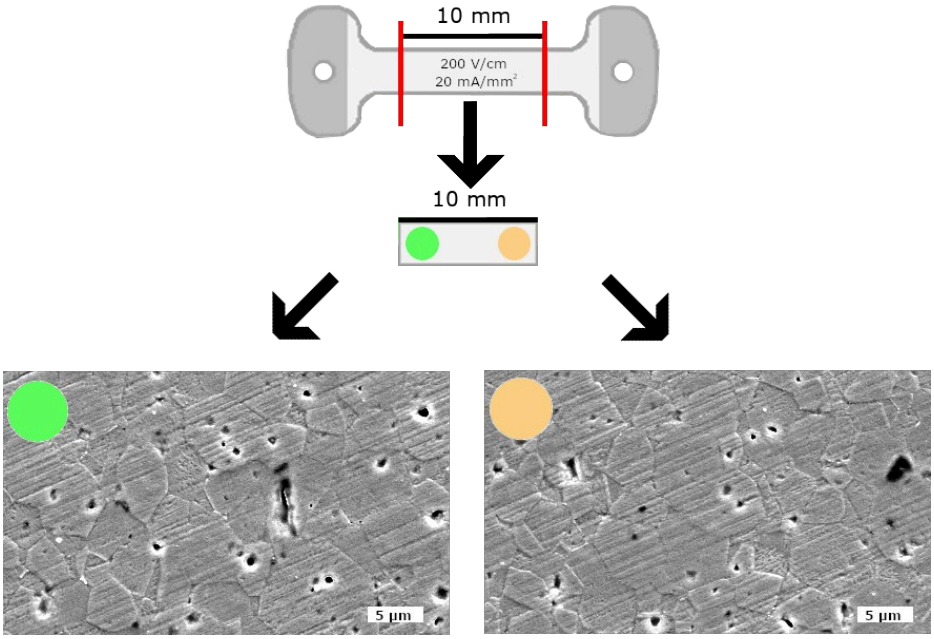

Fig. S4. Schematic representation of the sintered specimen ("dog bone" shaped) and the final sample after cutting, removing the electrodes zone. Below, SEM micrograph of two extreme zones of a final sample is shown. Electrical conditions are shown in the "dog bone".

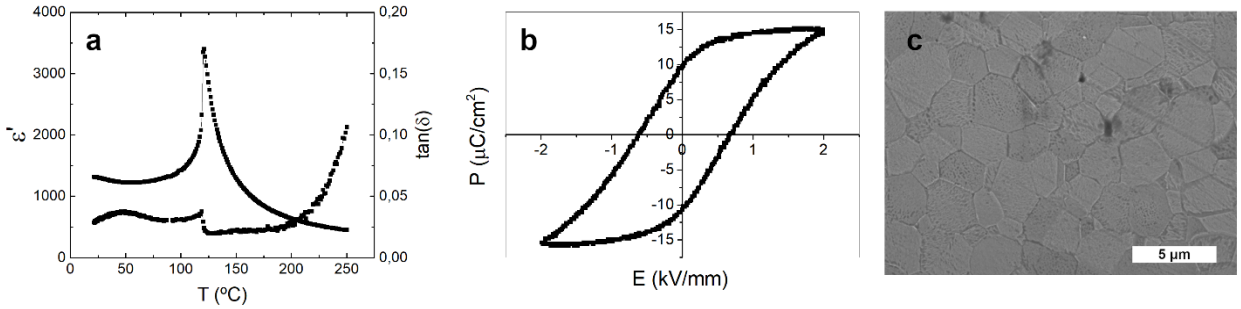

Fig. S5. (a) Temperature dependent dielectric permittivity, (b) ferroelectric hysteresis loop, and (c) microstructure of conventional sintered $BaTiO_3$.

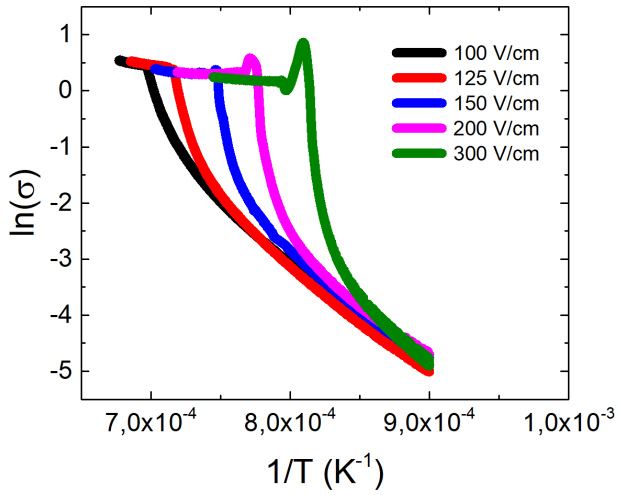

Fig. S6. Conductivity evolution of samples obtained in conventional flash sintering experiments carried out using different electric field values. Current density and dwell time are 15 mA/mm^2 and 10 min, respectively, for all experiments. Current spike is well-known as consequence of the change in the power supply operation mode.

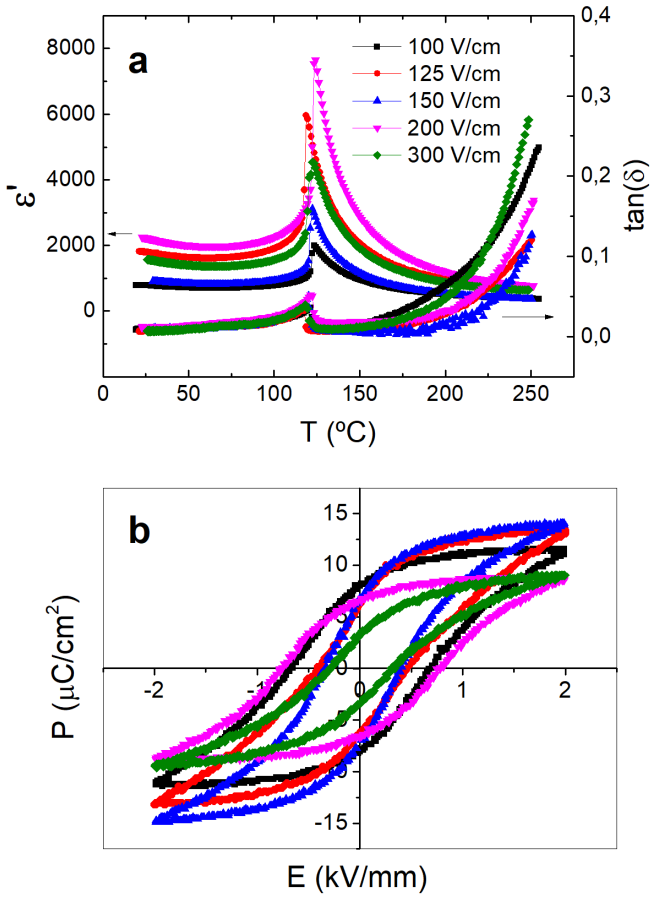

Fig. S7. (a) Dielectric and (b) ferroelectric responses of samples obtained in conventional flash sintering experiments carried out using different electric field values. Current density and dwell time have been maintained at 15 mA/mm^2 and 10 min, respectively.

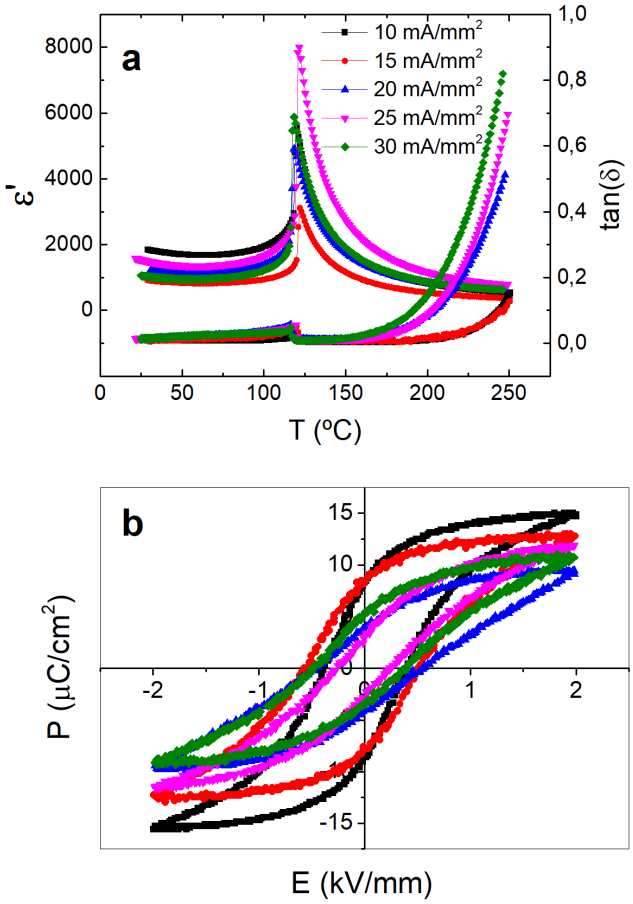

Fig. S8. (a) Dielectric and (b) ferroelectric responses of samples obtained in conventional flash sintering experiments carried out using different electric current density values. Electric field and dwell time have been maintained at 150 V/cm and 10 min, respectively.

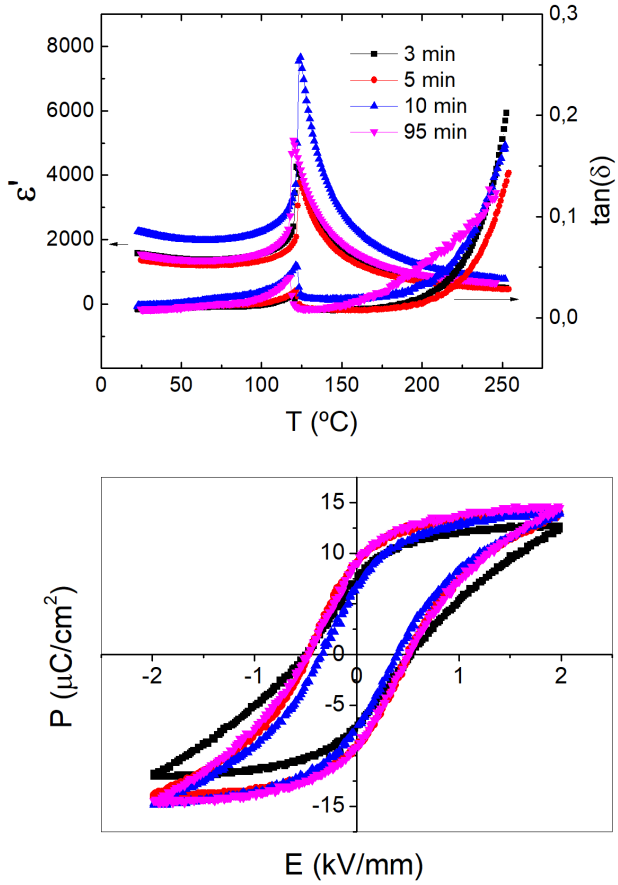

Fig. S9. (a) Dielectric and (b) ferroelectric responses of samples obtained in conventional flash sintering experiments carried out using different dwell times. Electric field and current density have been maintained at 200 V/cm and 15 mA/mm^2, respectively.

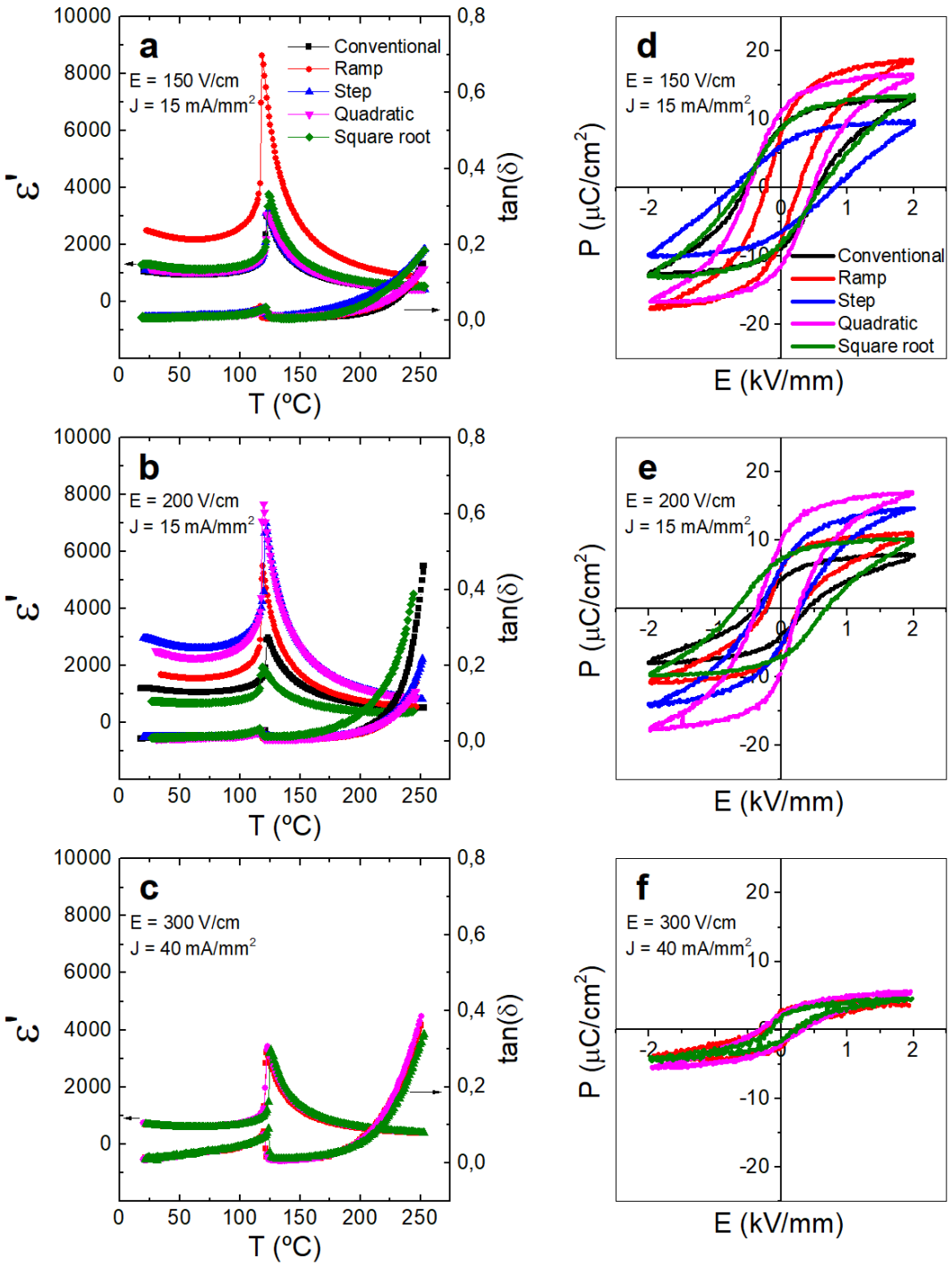

Fig. S10. (a, b, c) Dielectric and (d, e, f) ferroelectric responses of samples obtained in controlled current flash experiments carried out using different electric field and current density values. Results for samples with low density (< 90%) are not shown.

Table S1. Samples density (ρ) obtained in conventional flash and controlled current flash sintering experiments carried out using different electric current profiles. Dwell time has been maintained at 10 min.

E (V/cm)	J (mA/mm^2)	ρ (%)				
		Conventional Profile	Ramp Profile	Step Profile	Quadratic Profile	Square Root Profile
150	15	93,9	93,8	89,9	99,5	96,5
200	15	93,0	91,0	92,9	95,6	92,0
300	40	80,4	93,9	82,8	94,2	90,0