

Grain configuration effect on phase transition, piezoelectric strain and
temperature stability of KNN-based ceramics

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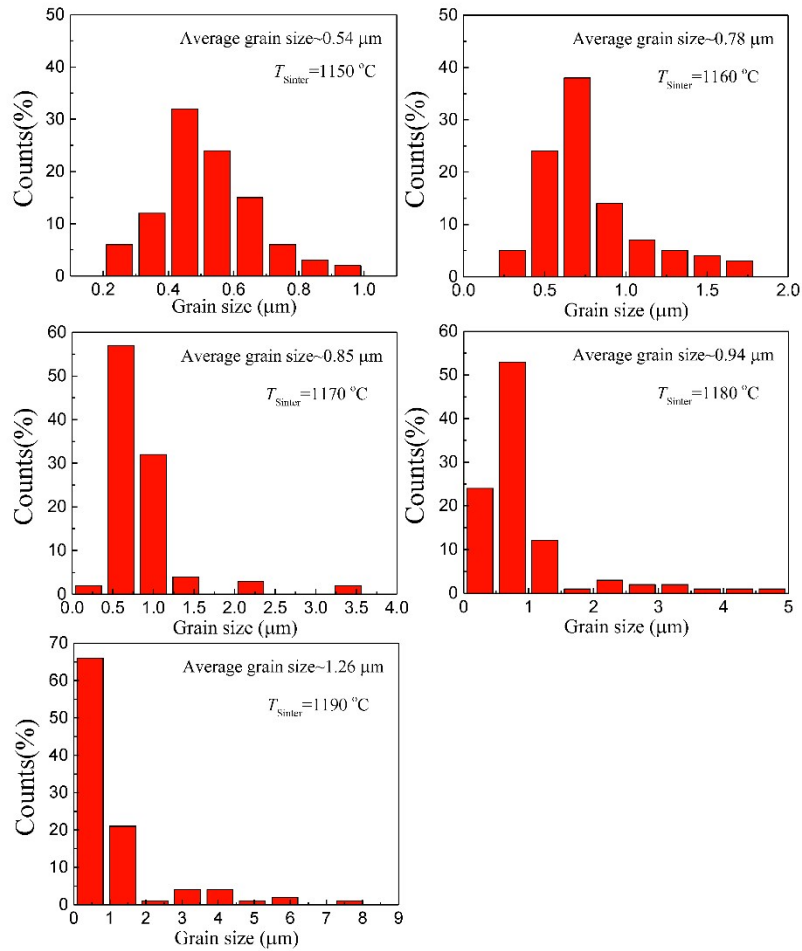


Fig. S1 Grain size distribution of 0.985(0.96KNNTa-0.04BNKZ)-0.015CZ ceramics sintered at different temperatures

The boundaries of the polygons with more than six sides are concave, and in the polygons with fewer than six sides the boundaries are convex. As grain boundaries migrate towards their centers of curvature, the grains with fewer than six sides tend to shrink, while the ones with more than six sides tend to grow.¹ Because the temperature field of the furnace is not the same, the calcined powder is difficult to be completely uniform. Thus, the side number of powder particle should be different. When T_{Sinter}

increases from 1150 °C to 1190 °C, powder particles with more than six sides grow at a higher rate and powder particles with fewer than six sides shrink at a higher rate. Thus, we consider that the smaller grains appear in samples sintered at 1170-1190 °C, which attributes to the shrinkage of powder particles with fewer than six sides. In other words, when T_{Sinter} increases from 1170 °C to 1190 °C, the small grains become smaller and coarse grains become larger. A similar report has been found.²

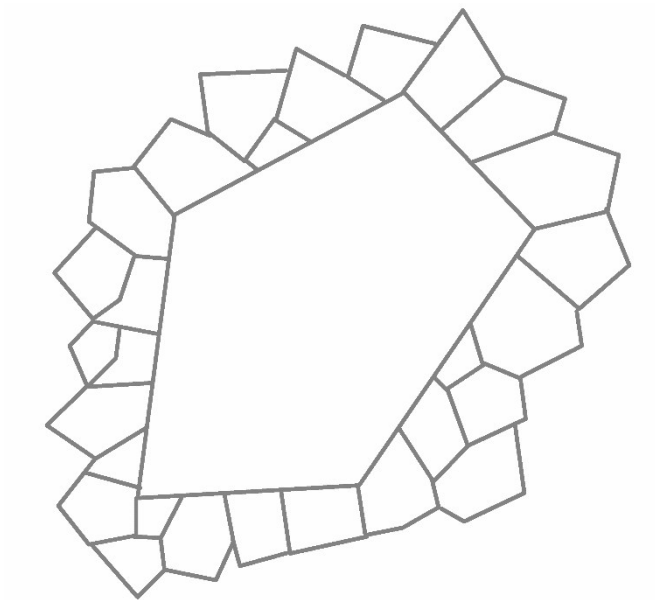


Fig. S2 Diagrammatic sketch of abnormal region

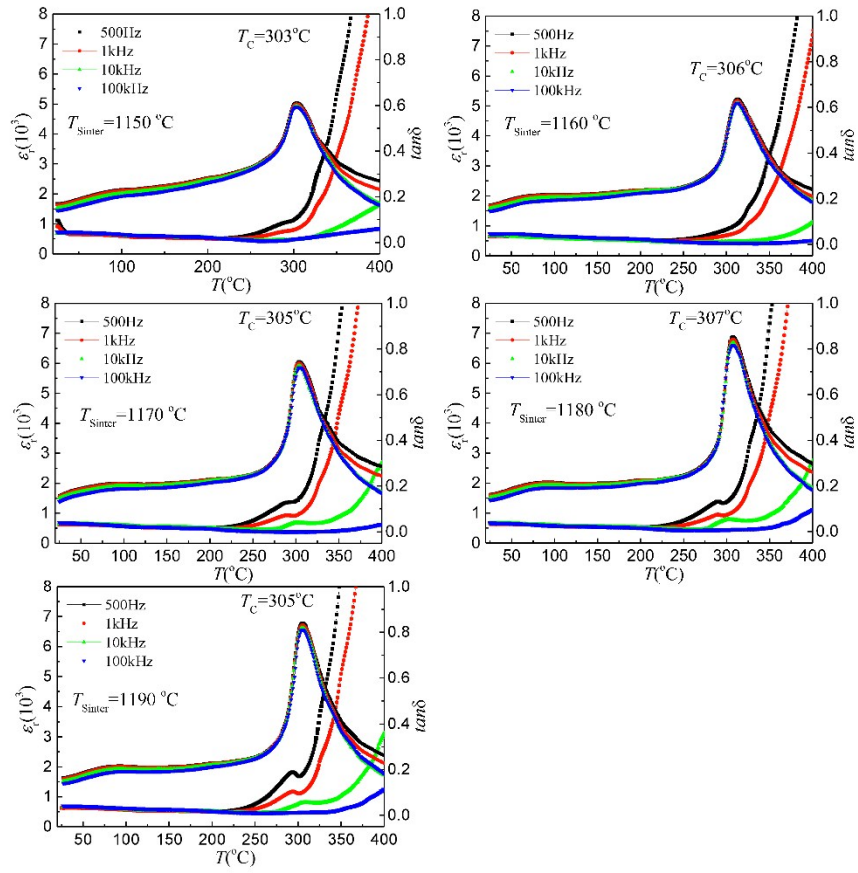


Fig. S3 Dielectric constant (ϵ_r) and dielectric loss ($\tan \delta$) as a function of temperature for 0.985(0.96KNNT-0.04BNKZ)-0.015CZ ceramics sintered at different temperatures

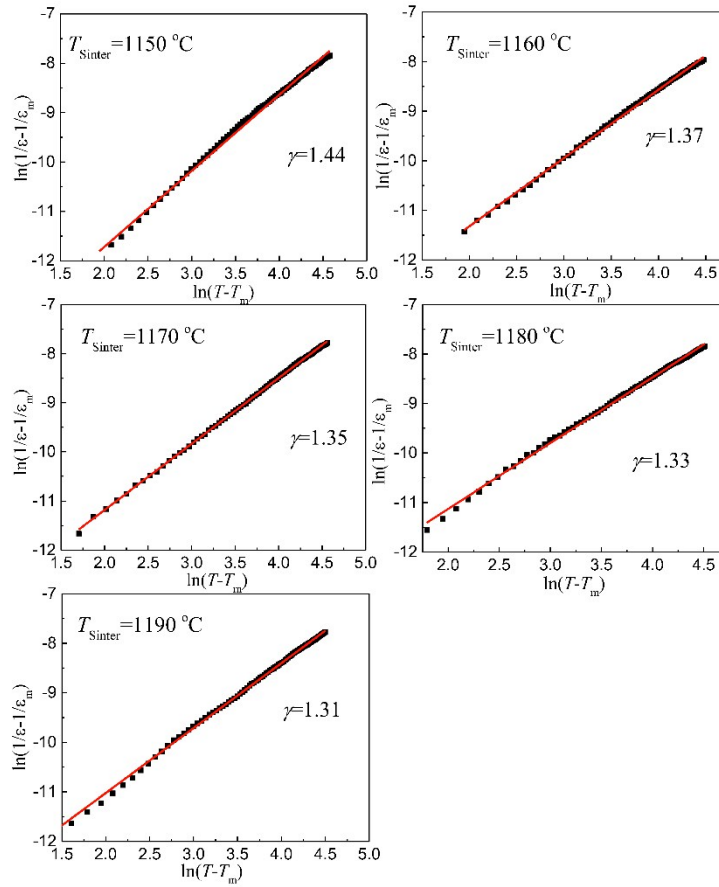


Fig. S4 $\ln(1/\epsilon - 1/\epsilon_m)$ as a function of $\ln(T - T_m)$ at 100 kHz for 0.985(0.96KNNT-0.04BNKZ)-0.015CZ ceramics sintered at different temperatures (Symbols: experimental data; Solid line: fitting to modified Curie-Weiss law)

1. Malič B, Koruza J, Hreščak J, Bernard J, Wang K, Fisher J, et al. Sintering of lead-free piezoelectric sodium potassium niobate ceramics. *Materials*. 2015;8(12):8117-8146
2. Amaral L, Fernandes M, Reaney I M, Harmer M P, Senos A M R, and Vilarinho P M. Grain growth anomaly and dielectric response in Ti-rich Strontium Titanate ceramics. *J Phys Chem C*. 2013;117(47):24787-24795