Stabilization of ferrielectric phase in NaNbO$_3$-based lead-free ceramics for wide-temperature large electrocaloric effect

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Fig. S1 Temperature dependent dielectric permittivity and dielectric loss at the frequency of 1 kHz-1 MHz for the NN-CZ ceramics. The inset is the evolution of $T_f$ with changing CZ content.

Fig. S2 Temperature-dependent XRD pattern of the NN sample.
Fig. S3 Temperature-dependent XRD pattern of the NN-0.01CZ sample.

Fig. S4 Temperature-dependent XRD pattern of the NN-0.02CZ sample.
Fig. S5 Temperature-dependent synchrotron XRD pattern of the NN-0.02CZ sample.

Fig. S6. Temperature-dependent Raman spectra of NN sample.
Fig. S7. Temperature-dependent Raman spectra of NN-0.02CZ sample.

Fig. S8. The corresponding intensity profiles of (1, 1-q, 1) superlattice reflections and (111) basic reflections for NN-xCZ ceramics.
Fig. S9 The proposed configuration for $q=1/4$ phase of NN ceramic.

Fig. S10 Temperature-dependent P-E hysteresis loops at 10 Hz of the NN-0.05CZ sample.
Fig. S11 Temperature dependence of the specific heat curve for the NN-0.05CZ ceramic.
The ECE (a) $\Delta S$ and (b) $\Delta T$ for the NN-0.05CZ sample under different applied fields, which exhibits excellent ECE compared with other lead-free ceramics.\textsuperscript{1-7}
Fig. S13 The temperature dependent recoverable energy storage density $W_{\text{rec}}$ and efficiency $\eta$ of the NN-0.05CZ ceramic.

$W_{\text{rec}}$ and $\eta$ can be calculated as follows:

$$W_{\text{rec}} = \int_{P_{\text{r}}}^{P_{\text{max}}} E \, dP$$

$$W_{\text{total}} = \int_{0}^{P_{\text{max}}} E \, dP$$

$$\eta = \frac{W_{\text{rec}}}{W_{\text{total}}}$$

where $P_{\text{max}}$, $P_{\text{r}}$, $E$ and $W_{\text{total}}$ are the maximum polarization, remanent polarization, applied electric field and total energy density.
References