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Figures

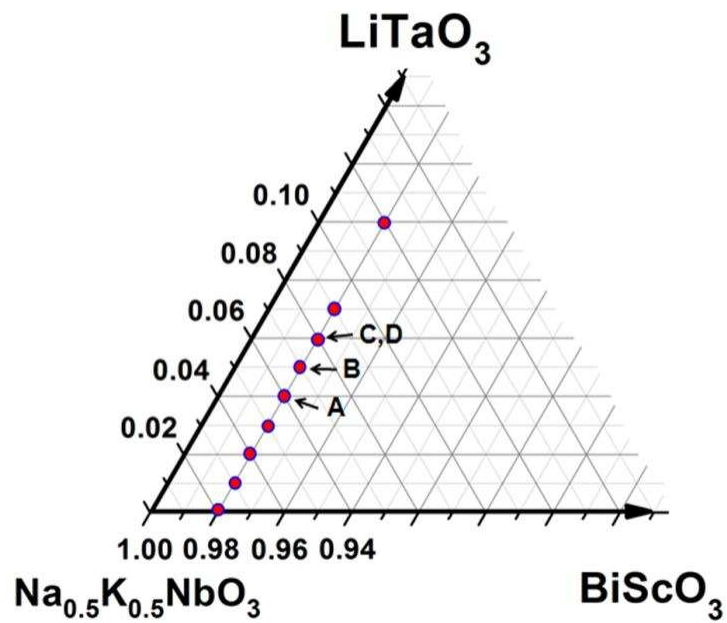


Fig. 1 Schematic diagram showing location of compositions in the NKN-xLT-2BS ternary system ($x=0-10$ mol%) with symbols A-D illustrating the compositions which were examined by TEM in this study, $x= 4-6$ mol% LT individually. Symbol D is the Excess 3 wt% alkali carbonates of NKN-6LT-2BS composition. Details can be obtained in **Table.1**.

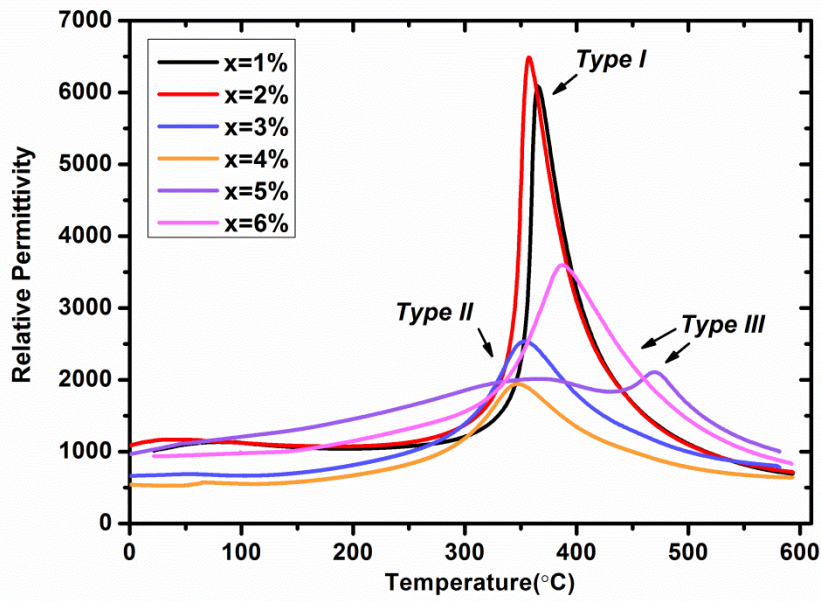


Fig. 2 Temperature dependence of relative permittivity at 100 kHz for NKN-xLT-2BS ceramics, $x=1-6$ mol% (a slight discontinuity at room-temperature is due to the use of a different instrument for measurements below room-temperature).

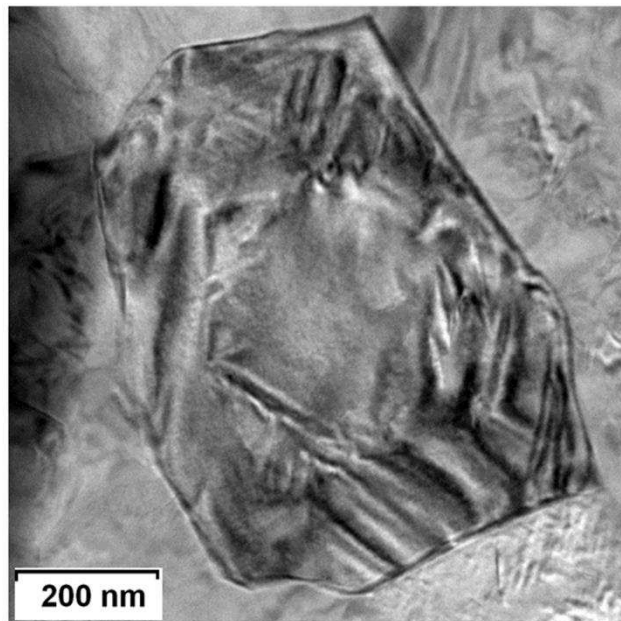


Fig. 3 Bright field TEM image for NKN-5LT-2BS specimen: clear evidence of core-shell structure grain is presented by the variations in diffraction contrast.

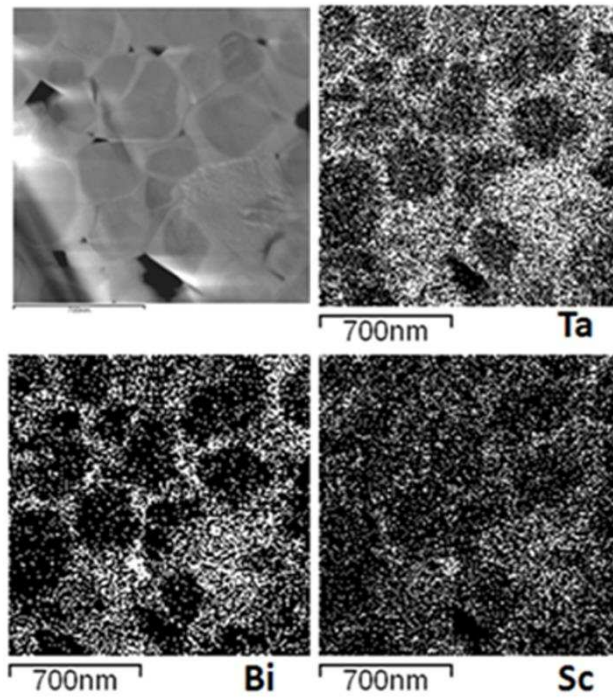


Fig. 4 STEM-HAADF image for NKN-5LT-2BS specimen (top left) and corresponding Sc($K\alpha$), Ta ($L\alpha$) and Bi ($L\alpha$) EDX elemental maps. Contrast in the STEM-HAADF image suggests chemical segregation within the grains, and is also confirmed by the elemental maps.

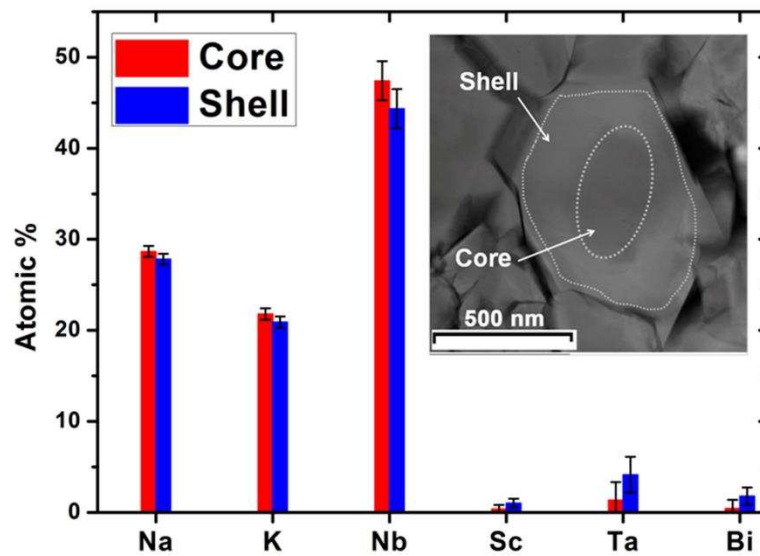


Fig. 5 NKN-5LT-2BS specimen: inset indicates example of core-shell grain. The

integrated EDX spectral analysis are presented in histogram form in main figure (and detailed in **Table. 2**)

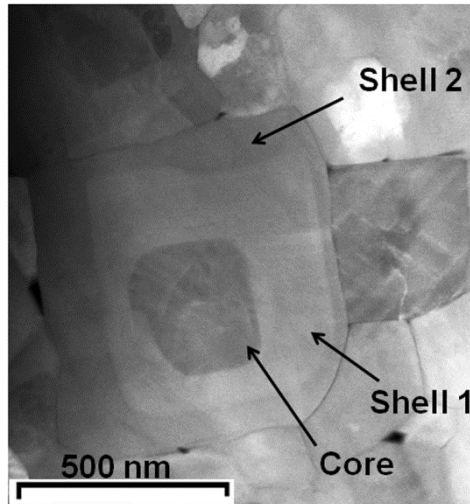


Fig. 6 NKN-6LT-2BS specimen: HAADF image showing three-tier core-shell structure.

Arrows illustrated the three regions of different contrast within the grain.

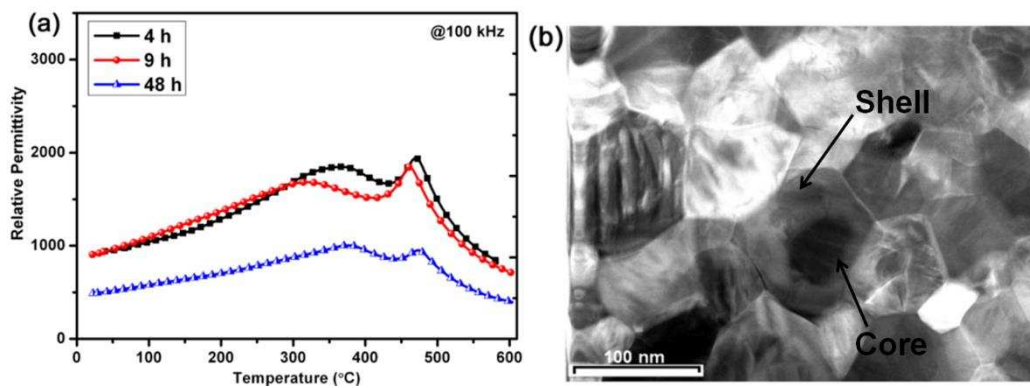


Fig. 7 (a) Relative permittivity versus temperature for NKN-5LT-2BS ceramics after different sintering times at 1100°C for 4, 9 and 48h; (b) TEM bright field image of NKN-5LT-2BS ceramic, after 48 h dwell time at 1100°C, indicating the specimen still has the core-shell structure.

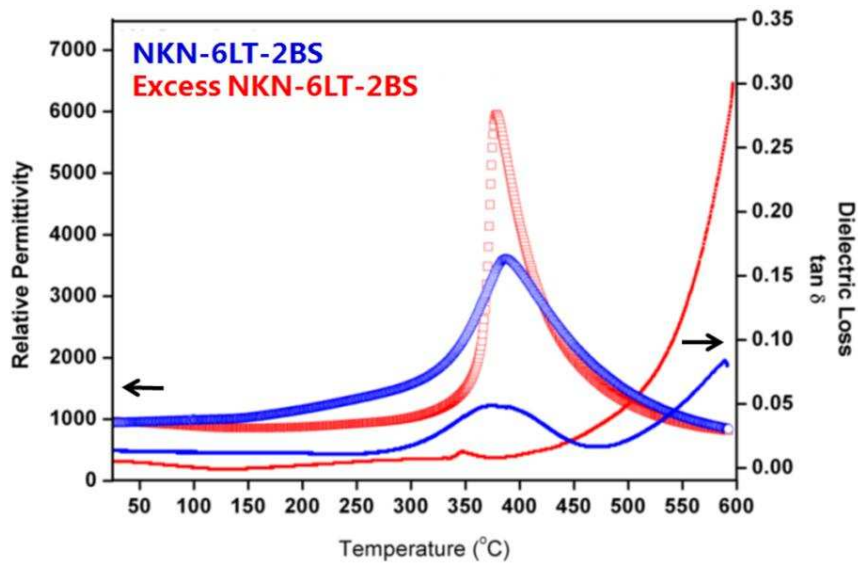


Fig. 8 Relative permittivity and dielectric loss versus temperature for standard and Excess NKN-6LT-2BS compositions.

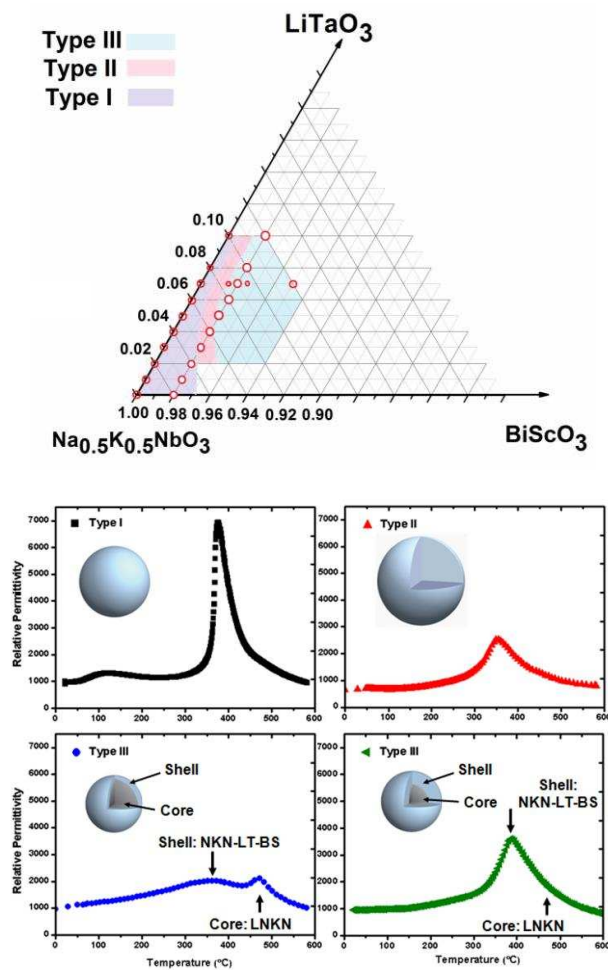


Fig. 9 Phase diagram of $\text{Na}_{0.5}\text{K}_{0.5}\text{NbO}_3\text{-LiTaO}_3\text{-BiScO}_3$ ternary solid-solutions, and summary plots of dielectric properties for sample Type I, II, and III (as described in main text). Schematic illustrations of the grain structures associated with each sample type are inserted in the plot.