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Thermal Evolution of Dielectric and Piezoelectric properties of Lead-Free Submicron-Structured $(Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}TiO_3$ Ceramics

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Abstract

The challenge to develop high piezoelectric sensitivity and lead-free compositions ferroelectric ceramics has bring new interest [1] to the study of some classical ferroelectrics as $(Bi_{0.5}Na_{0.5})TiO_3$ [2] and its solid solutions. The composition near the MPB of the system (1-x) $(Bi_{0.5}Na_{0.5})TiO_3$-$xBaTiO_3$ with x=0.06 (BNBT6) was found to have interesting properties as lead-free piezoelectric ceramic [3]. Processing of ceramics from nanopowders allows getting fine grained, submicron structured, ceramics that are of interest both for the basic studies of size-effects in ferroelectrics and for their use as high frequency ultrasonic transducers. Submicron-structured BNBT6 ceramics, obtained from nanometric powder synthesized by sol gel auto-combustion at 500ºC [4], by hot-pressing at low temperature (700-800ºC) and subsequent recrystallization at higher temperature, still moderate (<1100ºC), in order to reduce loss of the volatile elements, have been studied. Elastic and piezoelectric coefficients, as well as electromechanical coupling factors, were determined at the resonances of, thickness poled, thin disks and shear plates [5]. The best room temperature piezoelectric coefficient obtained in these BNBT6 fine-grained (~1 µm) ceramics (d_{33}=148 pC.N^{-1}, d_{31}= -37 pC.N^{-1}, d_{15}=158 pC.N^{-1}, k_t= 40.4%, kp=26.8% and k_{15}=40.2%) can be compared with those reported for coarse-grained ceramics prepared at higher sintering temperatures. The thermal evolution of the dielectric permittivity, piezoelectric coefficients and coupling factors has been also determined and compared with results reported for BNBT6 coarse grained ceramics [6], which showed a depolarization temperature of ~105ºC.

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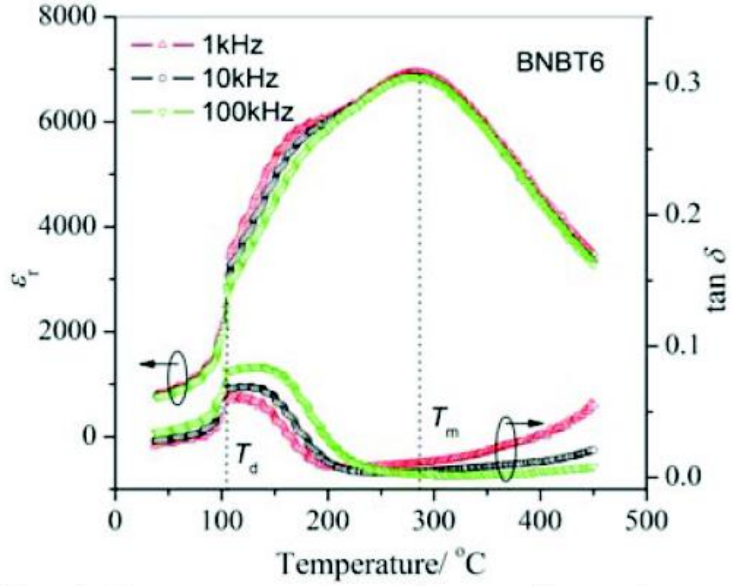


Fig. 1 Temperature dependence of ε_r and tan δ of the poled BNBT6 ceramics at 1, 10, and 100 kHz respectively.

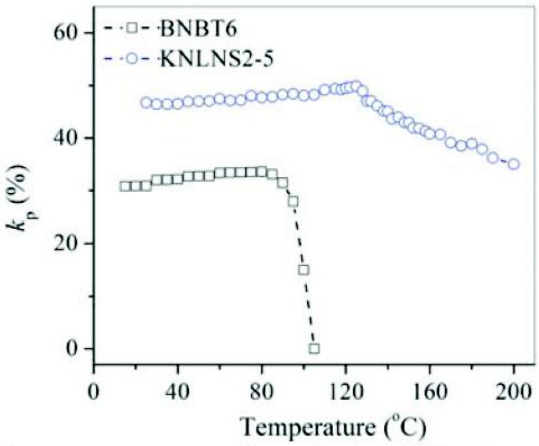


Fig. 3 Temperature dependence of k_p of BNBT6 and KNLNS2−5 ceramics.

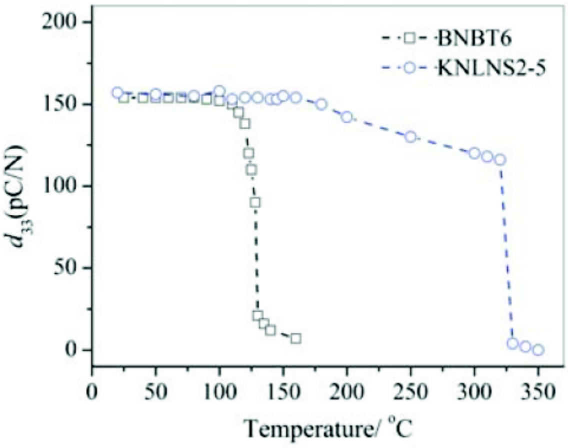
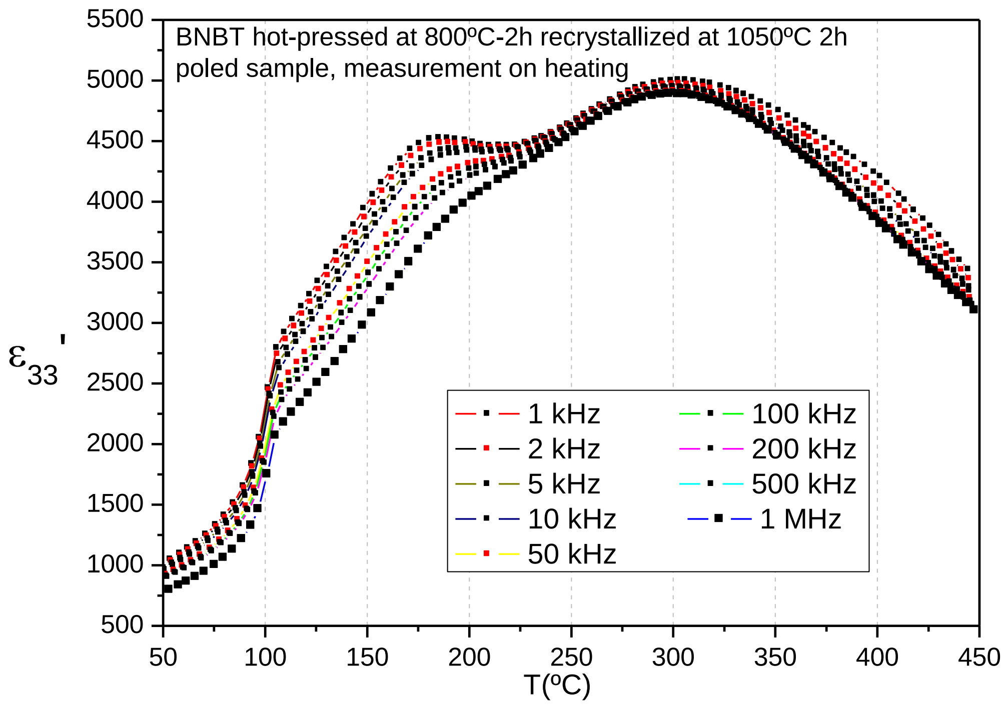
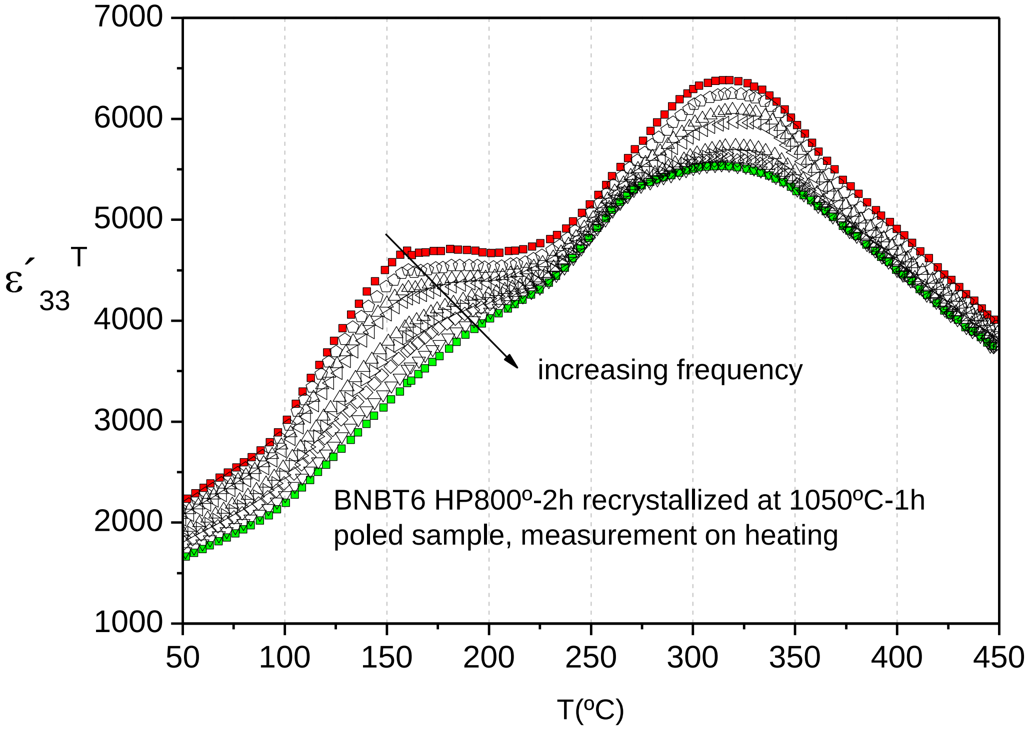


Fig. 4 Annealing temperature dependence of d_{33} of BNBT6 and KNLNS2−5 ceramics.

Figures From : D.Q. Xiao, L. Wu, J.G. Zhu. “Temperature stability of Lead-free Piezoelectric Ceramics of Perovskite $Bi_{0.5}Na_{0.5}TiO_3$ and $K_{0.5}Na_{0.5}NbO_3$ Families”. 18th IEEE International Symposium on the Applications of Ferroelectrics (ISAF2009) 3-27 Aug. 2009. Xiam, China. DOI:10.1109/ISAF.2009.5307596

From this work: the shape of the permittivity curve changes and, while piezoelectric properties are still high (see results reported in paper submitted to PAC journal), the depolarization temperature increases as the grain size decreases from sample recrystallised at 1050ºC-2h to sample recrystallised at 1050ºC-1h. For further decrease of the grain size, the temperature of depolarization remains similar but the piezoelectric properties are lower.





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Composition and synthesis route	BNBT6 sol-gel autocombustion					BNBT6[2] mixed oxides	BNBT6[3] mixed oxides	BNBT6[4] mixed oxides
Processing conditions	HP 700ºC2h 1000ºC 1h	HP 800ºC2h 1000ºC 1h	HP 800ºC2h 1050ºC 1h	HP800ºC2h 1050ºC 2h	sintering[1] 1100ºC 2h	Sintering 1150ºC 2h	sintering 1200ºC 2h	
Grain size	<1µm	~1µm	~1µm	≥1µm	≥1µm	>3µm	>3µm	---
Poling conditions	40 kV.cm^{-1} 180ºC	40-60 kV.cm^{-1} 180ºC	40-60 kV.cm^{-1} 180ºC	¿?	¿?		30 kV.cm^{-1} 60ºC	30–40 kV.cm^{-1} 80ºC
d_{33} (10^{-12}C.N^{-1})*	105	143	148	167	125	131	125	122
d_{31} (10^{-12} C.N^{-1}) P	-19.9+0.97i	-34.8 + 1.37i	-37.0 + 1.33i	-38	-39	-36.8	40	---
K_p(%) P	13.6	24.6	26.8	25.2	27.2	26.5	20	29
K_t(%) T	29.5	36.4	40.4	42.3	42.6	50.4	52	40
N_p(kHz.mm) P	2535	2873	2933	2995	2953	2937	2975	3000
N_t(kHz.mm) T	1855	2238	2281	2274	2409	2293	2600	2522
ε^T_{33} P	465-24i	636 – 34i	641 – 31i	775 (1kHz)	698 (1kHz)	648 (1kHz)	580 (1kHz)	601 (1kHz)
tan δ P	0.052	0.054	0.048	0.036 (1kHz)	0.030 (1kHz)	0.023 (1kHz)	0.013 (1kHz)	0.018(1kHz)
s^E_{11} (10^{-12} m^{2}N^{-1}) P	14.3 - 0.20 i	9.57 - 0.09i	9.73 – 0.07i	8.96	9.23	9.01	8.59	---
s^E_{12} (10^{-12} m^{2}N^{-1}) P	-3.89 + 0.05 i	-2.90 + 0.03i	-3.02 + 0.02i	-2.31	-2.69	-2.31	---	---

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