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Phase sequence and dielectric properties of $K_{0.5}Na_{0.5}NbO_3$ ceramics sintered by different methods

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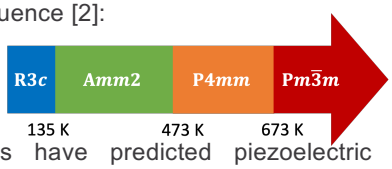
Summary

In this work, we revisit the phase transition sequence and the effect of the sintering process on the structure, lattice dynamics, and dielectric properties of KNN ceramics prepared by conventional sintering, spark plasma sintering, and spark plasma texturing.

Motivation

To substitute PZT-based materials, lead-free materials have been proposed, like $K_{0.5}Na_{0.5}NbO_3$ (KNN)[1], having the following phase sequence [2]:



Theoretical calculations have predicted piezoelectric response enhancement when T_{T-C} become closer to T_{O-T} [3], in which sintering conditions could play an important role [4,5]:

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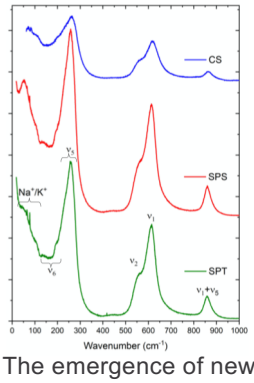
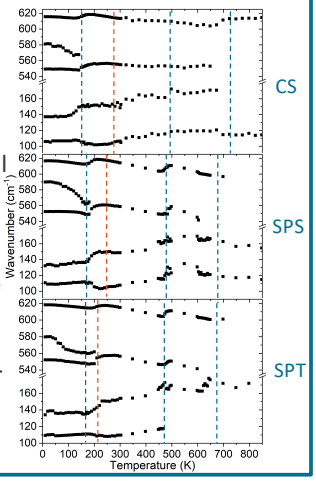
Sintering method	d_{33} (pC/N)	Grain size(μm)	Relative density (%)	Internal stress (MPa)
Conventional sintering	CS	110	3.6	95.3	0
Spark Plasma sintering	SPS	50	3.0	96.0	32
Spark Plasma Texturing	SPT	125	1.4	99.8	108

Lattice dynamics

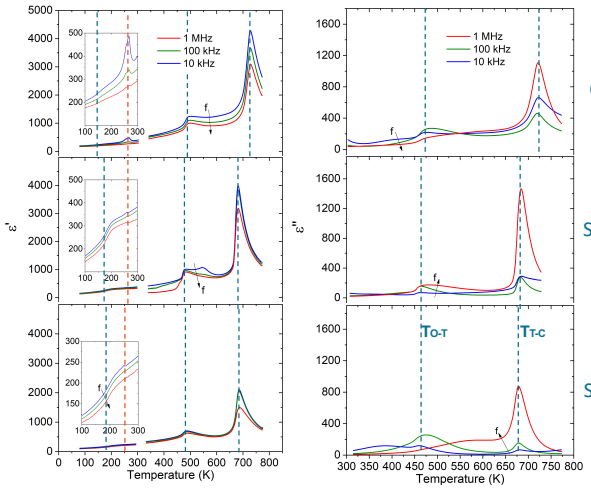


Similar room temperature spectra reveal identical crystallographic structure and chemical composition of the samples .

The emergence of new Raman bands and/or the abrupt changes in the temperature dependence of their wavenumber indicates structural changes associated with phase transitions (blue dash-lines). In the low temperature regime, an additional smooth change in the wavenumber can be observed, marked with an orange dash-line. This probably points out for a new intermediate phase (X-phase).



Dielectric Permittivity



The frequency independent anomalies:

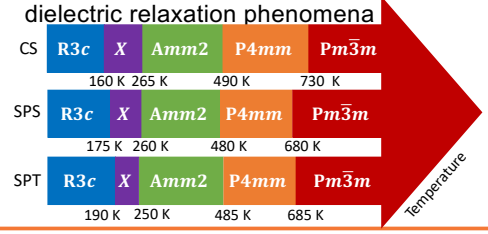
- mark phase transitions, where two transitions can be observed in the low temperature regime, corroborating the existence of the X-phase.
- At 100 kHz, ε′(T) maximum is similar for the CS and SPS samples, and half of the value for SPT. This could be associated with the smaller grain size of this sample.

The frequency dependent anomalies:

- more expressive near T_{O-T} for the SPT sample, mirrors dielectric relaxational process.

Conclusions

- Independently on the sintering method:
  - ✓ Phase transition sequence includes an unreported structural and polar phase at low temperature
- Dependently on the sintering method:
  - ✓ Apparent changes of the stability temperature interval of the different phases.
  - ✓ The dielectric strength and emergence of dielectric relaxation phenomena



References:

[1] I. Coondoo et al., J. Advanced Dielectrics, 03, 1330002 (2013)
[2] B. Orayech et al., J. Appl. Cryst., 48, 318-333 (2015)
[3] D. Damjanovic, Appl. Physics Letters, 97, 062906 (2010)
[4] R. Pinho et al., Appl. Mater. Today 19, 100566 (2020)
[5] M.M. Gomes et al., Ceram. Int., 47, 8308-8314 (2021)

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