Accepted Manuscript

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PII: S0925-8388(18)31774-2

DOI: 10.1016/j.jallcom.2018.05.089

Reference: JALCOM 46062

To appear in: *Journal of Alloys and Compounds*

Received Date: 1 January 2018

Revised Date: 5 May 2018

Accepted Date: 7 May 2018

Journal of Alloys and Compounds

Structural evolution and dielectric properties of Nd and Mn co-doped $BaTiO_3$ ceramics

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ABSTRACT

$(Ba_{1-x}Nd_x)(Ti_{0.97}Mn_{0.03})O_3$ (BNTM) (x = 0.01, 0.02, 0.04, 0.06) ceramics were prepared using a conventional cold-pressing ceramic technique. The structure, valence state and dielectric properties were investigated using XRD, RS, SEM, TEM, EPR, and dielectric temperature and frequency measurements. XRD, RS analysis coupled with SEM and TEM observations indicate that the samples have a coexistence of tetragonal and hexagonal phase at room temperature as $x \leq 0.02$ and become a single phase in the tetragonal as $x = 0.04$ and in the cubic as $x = 0.06$ (air-sintered, 1400 ºC/12 h). It reveals that Nd^{3+} ions can suppress hexagonal phase effectively and benefit the formation of single-phase ceramics. Improvement of dielectric properties is accompanied by the structural evolution. Particularly a cubic ceramic with $x = 0.06$, the dielectric-peak temperature is found to shift to room temperature, meeting the EIA Y5V specification with tan $\delta < 0.04$. Up to $x = 0.04$, the phase transition remains first order. The valence state of Mn ions was analyzed by EPR. It is found that Mn ions transform from high valence (+3 and +4) to low valence (+2) with the increase of Nd concentration , and all of the Mn ions exist as Mn^{2+} when $x = 0.06$. The unit cell volume and dielectric-peak temperature of BNTM decrease nonlinearly with increasing x, which can be ascribed to the incorporation of Nd ions and the formation of $2Nd_{Ba}^{\bullet} - Mn_{Ti}''$ donor-acceptor defect complexes.

Key-words: $BaTiO_3$; Structural evolution; Valence state; Defect complexes; Dielectric properties; Electron paramagnetic resonance

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