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Texturization of potassium sodium niobate (KNN) ceramics in the presence of CuO and MnO

Pinho R.*, Asif M.*, Costa M.E*, Vilarinho P.M.*

*Department of Materials and Ceramic Engineering, Centre for Research in Ceramics and Composite Materials, CICECO, University of Aveiro, 3810-193 Aveiro, Portugal

Corresponding author: paula.vilarinho@ua.pt

Alkaline niobates, namely potassium sodium niobate (KNN) are currently considered promising lead free substitutes of some of the most important ferroelectrics / piezoelectrics, as $PbZr_{1-x}Ti_xO_3$ (PZT). High Curie temperature and relative high electromechanical coupling coefficients are important properties of KNN. However, properties of $KNbO_3$–$NaNbO_3$ compounds still need to be optimized. The use of rare earth dopants and texturization by Template Grain Growth (TGG) are some of the strategies currently under study. CuO and MnO have been used in KNN and others systems to improve densification and electrical properties. CuO as dopant improves densification and grain growth, that is attributed to liquid phase formation [1]. On the other hand doping with MnO enhances the densification, increases resistivity and reduces leakage current [2].

In this work a systematic study of the microstructure development of textured KNN based ceramics, in which CuO and MnO are used as sintering aids is carried out. (001)-oriented $K_{0.5}Nb_{0.5}NbO_3$ single crystals previously grown by high temperature flux method [3] were used as templates. KNN powders with 1.5% mol of CuO and 0.5% mol of MnO were mixed with 10 wt% of crushed single crystal. Pellets were pressed uniaxially at 170 MPa followed by cold isostatic pressing at 200 MPa. Sintering was carried out at 1065 °C for 2 h. The microstructure and the degree of orientation were characterized by scanning electron microscopy (SEM) and X-ray diffraction (XRD), respectively. The degree of crystallographic orientation was evaluated in terms of the Lotgering Factor (f) according to the equation: $f(00l) = \frac{P(00l) - P_0(00l)}{1 - P_0(00l)}$. Where, $P_{00l} = \sum I_{00l} / \sum I_{hkl}$ and $P_0 = \sum I^0_{00l} / \sum I^0_{hkl}$, with I_{hkl} and I^0_{hkl} being the intensities of (hkl) peaks for the textured and randomly oriented samples, respectively. f (00l) was calculated as ≈20% from the XRD patterns shown in Figure 1. Dense ceramics were obtained with a marked bimodal grain size distribution (Figure 2). The microstructure confirms the texturization of KNN by the growth of abnormal sized grains from the single crystal templates at the expense of the smaller grains, resulting in the (001) preferred orientation of the ceramics as observed by XRD. The textured KNN ceramics are composed of large grains with an average size of 60 μm and small matrix grains with an average grain size of 2.5 μm. Chemical elemental mapping by SEM EDX for K, Na, Nb,Mn, Cu and O performed on the surface of the ceramics (Figure 2) confirms the chemical homogeneity of the obtained textured ceramics.

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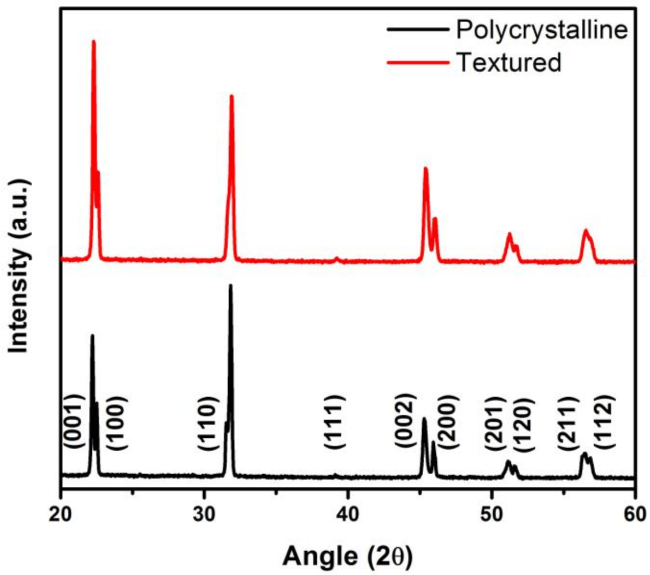

Figure 1: X-Ray diffraction patterns of polycrystalline KNN (bottom) and textured KNN (above) where it can be observed the difference in the intensities of peaks (00l).

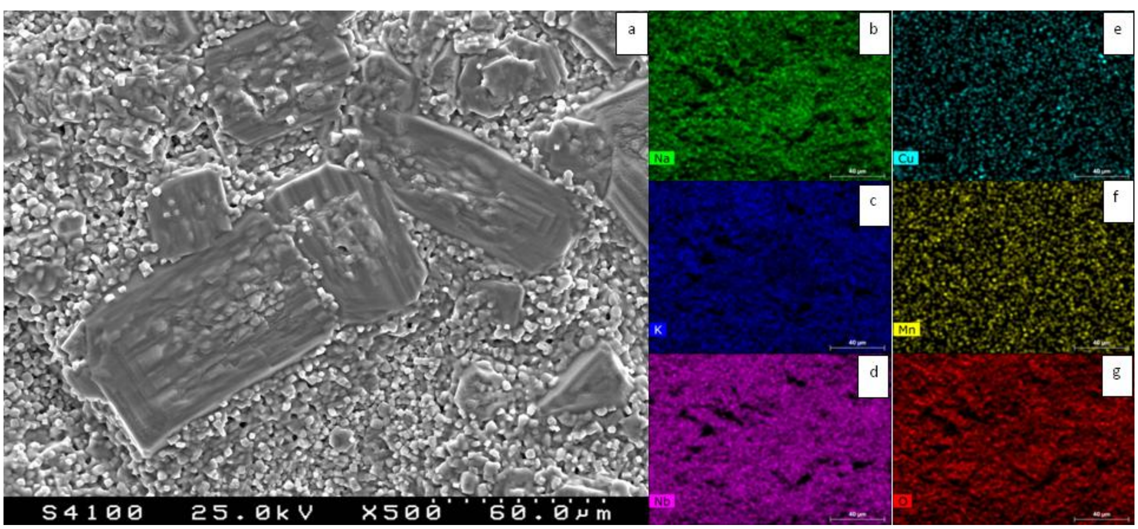

Figure 2: Microstructures of KNN+CuO+MnO (with 1,5% mol of CuO e 0,5% mol of MnO) sintered at 1065 ºC for 2 h: SEM image (a) and SEM EDX elemental maps for Na, K, Nb, Cu, Mn and O, respectively b, c, d, e, f, g). The textured KNN ceramics are composed of large grains with an average size of 60 μm and small matrix grains with an average grain size of 2.5 μm.

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