Elaboration and characterization of the tetragonal tungsten bronze ferroelectric $Sr_{2(1-x)}Na_{(1+x)}Gd_xNb_5O_{15}$

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Abstract

The nanopowders ferroelectric $Sr_{2(1-x)}Na_{(1+x)}Gd_xNb_5O_{15}$ was synthesized by solid-state method, were prepared by the precursor calcination at 1200 ℃ during 24 hours in oxygen atmosphere and by a high-temperature solid-state reaction technique. The structural characterization of $Sr_{2(1-x)}Na_{(1+x)}Gd_xNb_5O_{15}$ nanopowders was carried out by the X-ray diffraction. The profile of adjusting of the set of diffraction lines and refinement of the structural parameters were performed by the Rietveld method, using the FULLPROF program. The results reveal an orthorhombic phase, with space group Pmmm for $0 \leq x \leq 0.8$ and a tetragonal phase, with space group P4mm, for $0.9 \leq x \leq 1$. © 2017 ijrei.com. All rights reserved

Keywords: Solid-state, X-ray diffraction, Space group, Ferroelectric

1. Introduction

Niobates with a tetragonal tungsten bronze (TTB)-type structure are of great scientific, technical, and industrial interest as materials for laser modulation, frequency multiplicity, and the generation of second harmonics for applications in pyroelectric detectors and piezoelectric transducers [1]. The tetragonal tungsten bronze-type structure (TTB) can be considered as a derivative of the classical perovskite one. It can be described by the chemical formula $(A1)_2(A2)_4C_4Nb_{10}O_{30}$. A1, A2, and C denote different sites in the crystal structure. The A1 cavities have a cuboctahedral coordination of oxygen atoms, the A2 cavities a Penta capped pentagonal prismatic, and the C cavities a tricapped trigonal prismatic one. The size of these cavities decreases in the order A2 > A1 > C.

In this work, we report the growth and structural characterization of the new family of ferroelectric compounds of TTB type $Sr_{2(1-x)}Na_{(1+x)}Gd_xNb_5O_{15}$ (SNGN) as a function of x $(0 \leq x \leq 1)$. The aim of this work is the study of symmetry changes as a function of gadolinium (Gd^{3+}) concentration in SNGN compounds.

2. Sample preparation

Sodium strontium niobate, $Sr_{2(1-x)}Na_{(1+x)}Gd_xNb_5O_{15}$, was synthesized by solid-state method. Crystalline Nano powders were obtained by the precursor calcination at 1200℃ during 10 hours in oxygen atmosphere. The powder obtained from this calcination was characterized by X-ray diffraction (XRD). The diffraction pattern was refined in according to the Rietveld method. The refinement was performed using the program Full prof. The reason for having interest in the TTB family, it has been found that their physical properties can be modified by different ionic substitution at above mentioned sites [2]. The ferroelectric niobate materials with TTB structure are quite attractive materials due to their remarkable electro-optic, pyroelectric, piezoelectric, nonlinear optical properties [3].

3. Results and Discussion

Figure 1 shows the room temperature XRD patterns of $Sr_{2(1-x)}Na_{(1+x)}Gd_xNb_5O_{15}$ (SNGN) with x=0 (first compound) and x=1 (last compound). The patterns of two compositions indicate that the powders are pure and that they been have crystallized in a TTB-type structure. The intensity of some peak is weak for the sample SNN (x=0). After introducing of Gd^{3+} concentration in the sample SGN (x=1), the intensity of all peaks increases, suggesting that some distortion of crystal structure appears in Nb-sites. The diffraction patterns of the samples were refined according to the Reitveld method, using the Fullprof program integrated in WINPLOTR software, to determine the symmetry group and the parameters (a, b and c) for three compositions (figure 2).
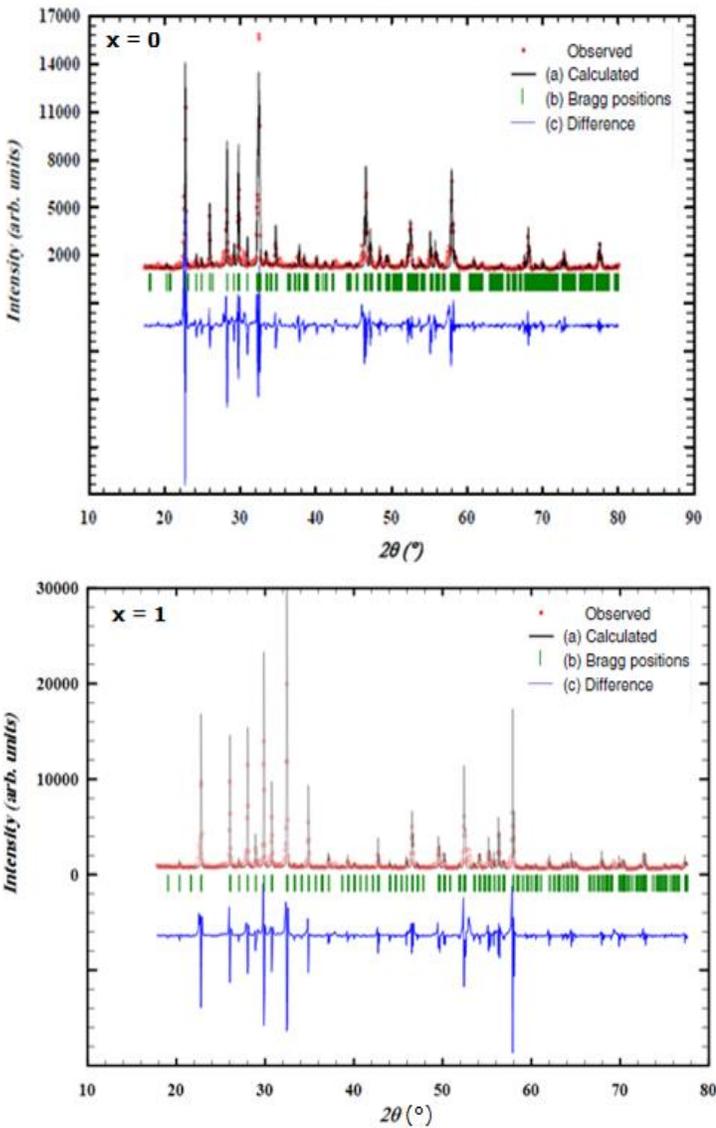

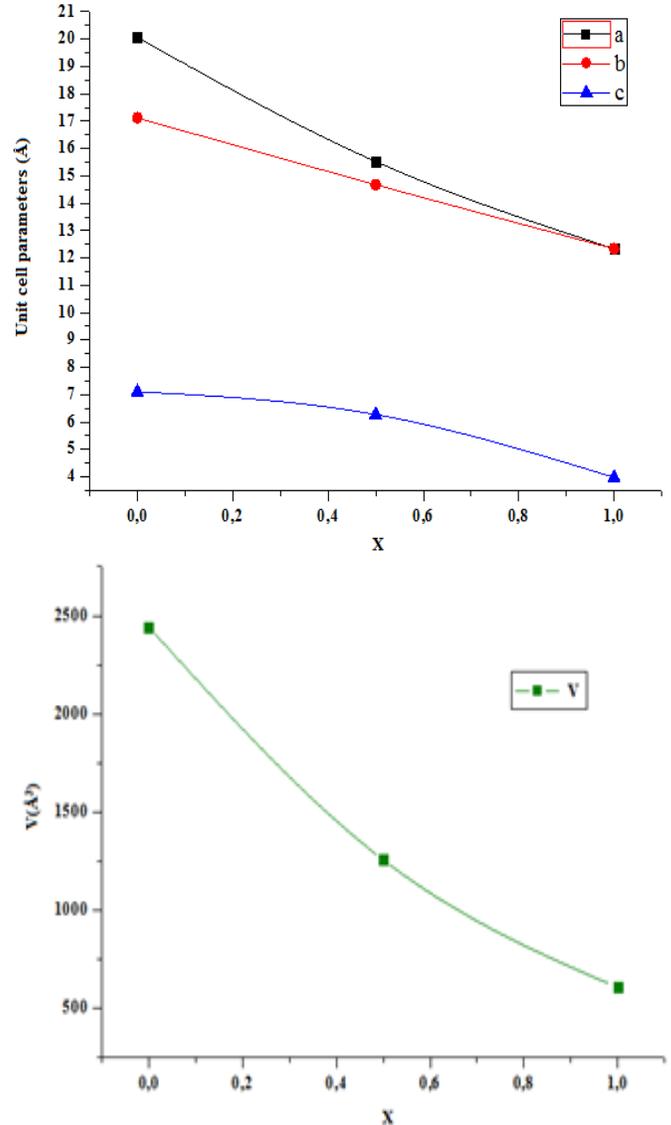*Figure 1: Observed, calculated and difference X-ray diffraction patterns for compounds with x=0 and x=1*

Figure 2: Evolution of the lattice parameters of $Sr_{2(1-x)}Na_{(1+x)}Gd_xNb_5O_{15}$ as a function of the gadolinium content x.

Figure 2 shows the presence of an orthorhombic phase, with space group Pmmm for $0 \leq x \leq 0.8$ and a tetragonal phase, with space group P4mm for $0.9 \leq x \leq 1$. Consistent with the work reported for x = 1 by Neurgaonkar et al.[4]. In the orthorhombic Pmmm phase, the polar axis lies in the ab-plane like in SNN. While in the tetragonal P4mm phases, the polar axis is along the c-axis. These results suggest a rotation of the polarization of 90° across the phase boundary, which separates the orthorhombic and tetragonal phases. Therefore, the rotation of the polarization can be favored around the phase boundary near x = 0.8. An enhancement of physical properties can be considered near this concentration of Gd^{3+}, as in the case of some solid solution ferroelectric perovskites: $PbZr_{1-y}Ti_yO_3$, $Pb[Mg_{1/3}Nb_{2/3}]_{1-x}Ti_xO_3$, and $Pb[Zn_{1/3}Nb_{2/3}]_{1-x}Ti_xO_3$ [5-6].

Concerning the lattice parameters (a, b and c) for each composition (figure 2) we note that when the substitution rate

x increases the parameters (a) and (b) decreases while the parameter (c) remains relatively constant before undergoing significant decrease for the large values of the substitution rate (x). Indeed, more the rate of gadolinium cation (Gd^{3+}) is important in the crystal lattice, the cell volume decreases. The shrinkage of the unit cell can be reasonably attributed to the smaller ionic size of Gd^{3+} (r = 1.02 Å) compared to that of Sr^{2+} (r = 1.18 Å) and equal to Na^+ (r = 1.18 Å).

4. Summary and Conclusion

We prepared a series of new family of TTB ferroelectric Materials $Sr_{2(1-x)} Na_{(1+x)} GdxNb5O15$ with $0 \leq x \leq 1$ using a solid-state method. The X-ray diffraction indicates that the powders are pure and crystallize in a TTB-type structure. We have successfully refined the diffraction patterns and determined the lattice parameters of each composition. Our results shows two ferroelectric phases: an orthorhombic phase with space group Pmmm for $0 \leq x \leq 0.8$ and a tetragonal phase, with space group P4mm, for $0.9 \leq x \leq 1$.

Moreover, we concluded than more the substitution rate x is big more the cell volume is small.

This work anticipates our future investigation of the ferroelectric TTB family

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