

NOVEL (K,Na)NbO₃ BASED PIEZOELECTRIC CERAMIC

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Abstract

Novel (1-x)(K_{0,5}Na_{0,5})NbO₃ – xGdAlO₃ piezoelectric ceramics were obtained using solid state method. The phase purity was confirmed by X-ray diffraction. The unit cell parameters were refined. The results show the presence of a phase transition from orthorhombic to tetragonal.

Introduction

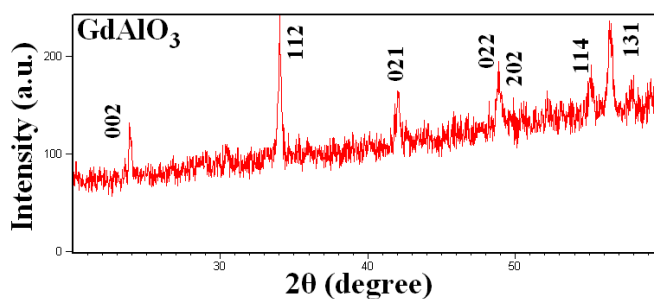
(K_{0,5}Na_{0,5})NbO₃ ferroelectric ceramics constitute an environmental friendly substitute to Pb(Zr, Ti)O₃ (1). The applications of such materials are capacitors, sensors, transducers or piezoelectric actuators (2, 3). As shown previously, the improvement of the piezoelectric properties of (K_{0,5}Na_{0,5})NbO₃ based ceramics can be accomplished through chemical substitution [4], structural material modification [5] or phase transitions manipulation [6]. In this paper we are considering the use of a new dopant, namely GdAlO₃, in order to change the crystalline structure from orthorhombic to tetragonal, since the presence of such phase transition can lead to serious improvements of the piezoelectric properties [7].

Experimental

(1-x)(K_{0,5}Na_{0,5})NbO₃ – xGdAlO₃ (noted KNN – xGAl), with x= 0 mol%, 0,25mol%, 0,75mol%, 1 mol%, 2,5 mol% and 5 mol% were obtained using the solid state method in air. The raw materials were K₂CO₃ (99%; Scharlau, Sentmenat, Spain), Na₂CO₃ (99%; Scharlau), Nb₂O₅ (99%, Merck, Darmstadt, Spain), Gd₂O₃ (99%; Fluka, Buchs, Switzerland) and AlN₃O₉•9H₂O (99%, Fluka, Buchs, Switzerland). First the dopant material was prepared in air at 1100°C. Then the piezoelectric powders were heated at 850°C for 6h, with a heating rate of 5 °/min. After calcinations, the powders obtained were grinded and mixed with a 5 mass% polyvinyl alcohol binder solution. Disk samples of 6 mm in diameter and approximately 1 mm thick were cold pressed at 200Mpa. The disks were sintered at 1050°C for 6h. After grinding, the crystalline structure of the sintered samples was examined by x-ray diffraction using a PanAnalytical X'Pert Pro MPD diffractometer. The refinement of the unit cell was performed using Treor algorithm in X'Pert HighScore Plus software,

Results and discussion

The experimental conditions used were sufficient to obtain pure GdAlO₃ powders, as we can observe in figure 1. The pattern was identified using the JCPDS-ICDD file number 00-009-0085: crystal system: orthorhombic, space group Pbnm (62), a= 5.2470 Å, b= 5.3043 Å, c= 7.4470 Å; α,β,γ= 90°; volume of unit cell: 207.25 Å³.

Figure 1. X-ray diffraction pattern of GdAlO_3 .

The X-ray diffraction pattern presented in figure 2 confirmed that the reference sample KNN is indexed as perovskite with an orthorhombic crystalline structure (space group $P2mm(25)$). The key feature of the orthorhombic KNN is represented by the variations in intensity of the peaks splitting at $2\theta \approx 22^\circ$ and 45° , respectively (010)-(001) and (020)-(002) crystalline planes.

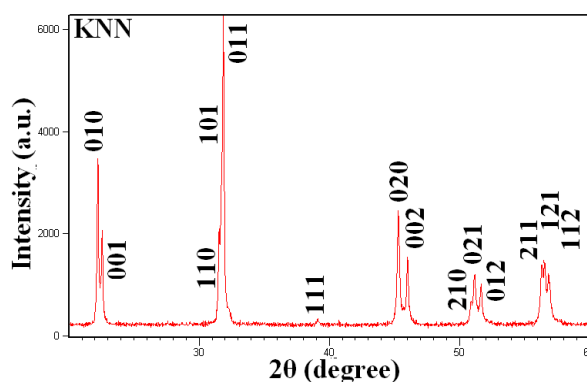
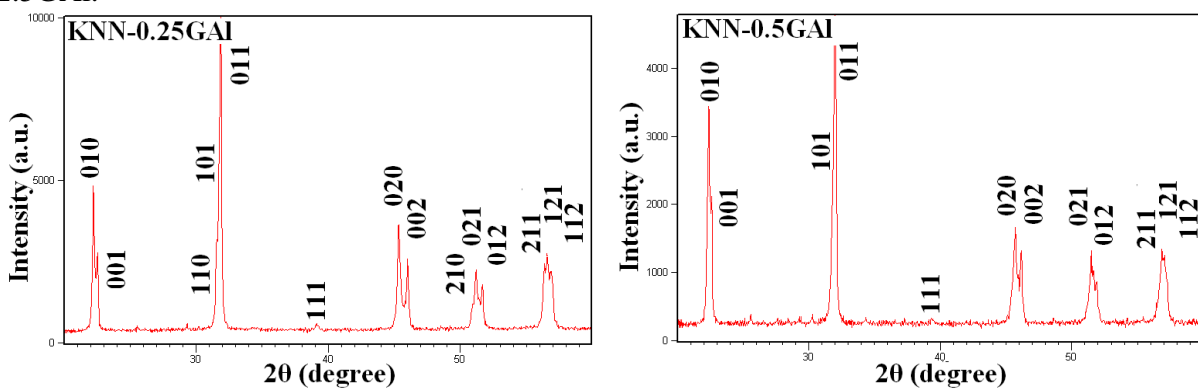


Figure 2. X-ray diffraction pattern of reference sample KNN.

In figure 3, all x-ray diffractions of KNN-GAl are presented. As previously discussed, we can observe a shifting in the peaks at $2\theta \approx 22^\circ$ and 45° , suggesting a shift in crystalline structure, from orthorhombic to tetragonal. Indeed, the refinement of the unit cell parameters show a shifting from $P2mm(25)$ orthorhombic to $P4mm(99)$ tetragonal starting from KNN-2.5GAl.



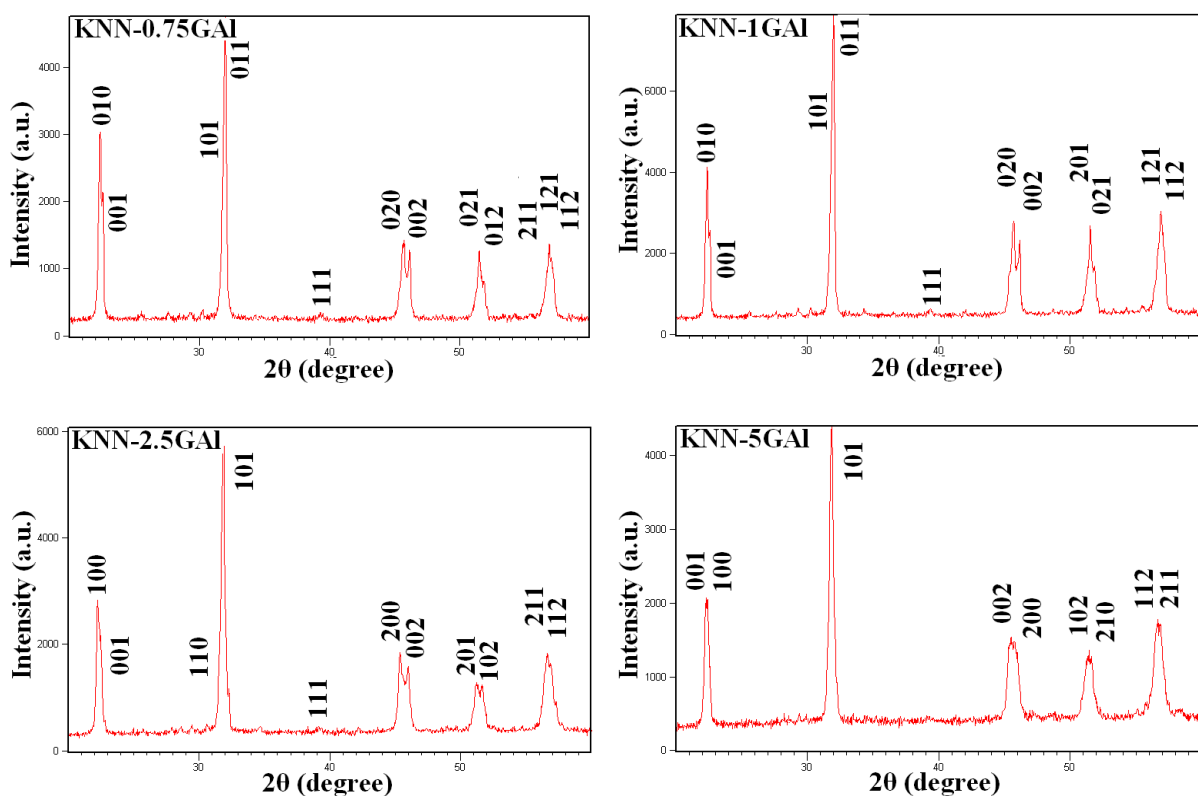


Figure 3. X-ray diffraction patterns of KNN-xGAl

The results of the unit cell refinement are presented in figure 4 and 5. As shown in figure 4, starting from the reference sample KNN, all cell parameters decrease. Above 2.5 mol% GdAlO₃, a=b since the unit cell is tetragonal.

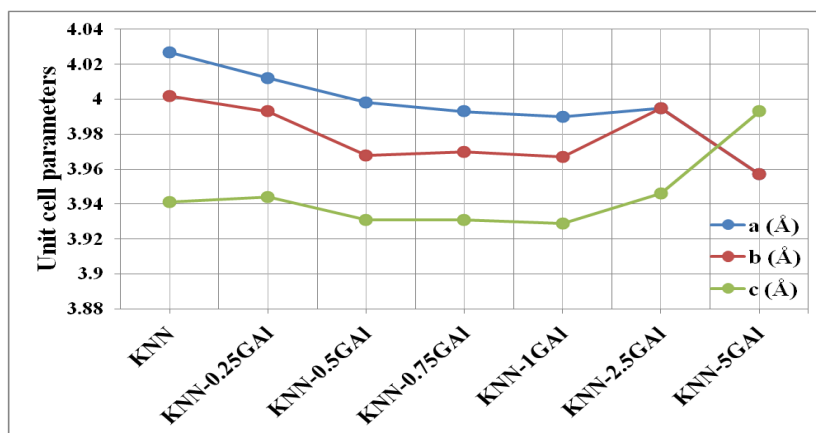


Figure 4. Unit cell parameters of KNN-xGAl

The unit cell volume variation depicted in figure 5 clearly shows the decrease of the unit cell in the orthorhombic phase, followed by a slight increase of the volume in the tetragonal phase. The abnormal variation at KNN-2.5GAl is to be attributed to the presence of a mixture of tetragonal and orthorhombic phases for this compound.

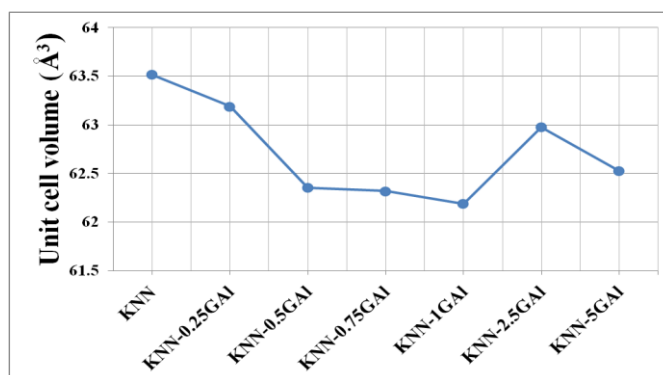


Figure 5. Unit cell volume of KNN-xGAl

Conclusion

$(1-x)(K_{0.5}Na_{0.5})NbO_3 - xGdAlO_3$ ceramics were obtained by solid state method. The addition of $GdAlO_3$ induce a phase transition, from orthorhombic to tetragonal at 2.5 mol $GdAlO_3$. Also the addition of $GdAlO_3$ tends to decrease the unit cell parameter, except for the tetragonal phases (KNN-2.5GAl and KNN-5GAl), where the unit cell parameters increase. The presence of a mixture of phases (orthorhombic and tetragonal) at 2.5 mol $GdAlO_3$, can be an indicator of the improvement of the piezoelectric properties.

Acknowledgements

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References

- [1] Li, J.F.; Wang, K.; Zhu, F.Y.; Cheng, L.Q.; Yao, F.Z., *J. Am. Ceram. Soc.* 2013, 96, 3677–3696.
- [2] Lee, T.; Kwok, K.W.; Li, H.L.; Chan, H.L.W, *Sens. Actuators A Phys.* 2009, 150, 267–271.
- [3] Shen, Z.Y.; Li, J.F.; Chen, R.M.; Zhou, Q.F.; Shung, K.K. *J. Am. Ceram. Soc.* 2011, 94, 1346–1349.
- [4] Lu, L.; Gong, Y.Q.; Gong, L.J.; Dong, H.D.; Dong, H.; Yi, X.F.; Zheng, X.J., *Mater. Des.* 2012, 33, 362–366.
- [5] Fu, J.; Zuo, R.Z.; Wang, X.H.; Li, L.T., *J. Alloys Compd.* 2009, 486, 790–794.
- [6] Toshio, K.; Yuan, Y.; Fumito, *Materials* 2010, 3, 4965–4978.
- [7] J. Rodel, W. Jo, K.T.P. Seifert, E.M. Anton, T. Granzow D. Damjanovic, *J. Am. Ceram. Soc.* 92 (2009), 1153.