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Effect of samarium concentration on the structural and electrical properties of (K, Na) NbO_3 thin films

Nurul Azuwa Azmi[1], Umar Al-Amani Azlan[2,*], Mohd Asyadi 'Azam Mohd Abid[1], Mohd Warikh Abd Rashid[1], Maziati Akmal Mohd Hatta[1]

[1)] Faculty of Manufacturing Engineering, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia
[2)] Faculty of Engineering Technology, Universiti Teknikal Malaysia Melaka, Hang Tuah Jaya, 76100 Durian Tunggal, Melaka, Malaysia

[*]Corresponding e-mail: umar@utem.edu.my

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ABSTRACT – Potassium sodium niobate have been chosen as a new candidate to replace the usage of lead-based materials due to its excellent properties. However, volatilities of K and Na can affect the electrical properties. This problem can be minimized by adding a doping element into A-site ions which is Samarium. A simple sol-gel route was used to produce the thin films with different dopant concentrations. In this work, the structural and electrical properties were determined by using XRD and LCR meter, respectively. It was found that the structure of KNN had a tendency to change from orthorhombic to tetragonal phase, thus resulting in better electrical properties of the KNN thin films.

1. INTRODUCTION

Recent studies on lead-based materials has led to the development of lead-free materials due to the environmental concern on the usage of lead-based materials especially the $PbZrTiO_3$ (PZT). The regulation have been made by the Restriction of Hazardous Substances (RoHS) due to its dangerous effect on human and other living things [1] [2]. Therefore, potassium sodium niobate, (K, Na) NbO_3 (KNN) has been a good candidate to replace the high usage of PZT materials. KNN has been recognized recently due to its high Curie temperature (420°C), high dielectric constant (~700), high remanent polarization (14 $\mu C \cdot cm^{-2}$), low coercive field (~140 $kV \cdot cm^{-1}$) and high piezoelectric constant [3]. Nevertheless, the volatilization of K and Na at certain temperature resulted in poor electrical properties of KNN, likely due to defects and oxygen vacancies [3][4]. Therefore, this problem can be minimized by doping of A-site ions, $(K_{0.5}, Na_{0.5})^+$ and/or B-site ion Nb^{5+} which happened to be improved its physical and electrical properties [5][6]. Hence, this present study is focusing on the addition of Samarium (Sm), for better electrical properties of KNN with different doping concentrations.

2. METHODOLOGY

Sodium acetate and potassium acetate was used as the starting precursors. Subsequently, samarium nitrate hexahydrate was added with different concentrations of 0.1, 0.3, 0.5, 0.7 and 0.9 mol % as the doping element and mixed into 2-methyloxyethanol. In the meantime, niobium ethoxide and acetyl acetone (chelating agent) were also mixed into 2-methyloxyethanol in different beaker. After continuous stirring for 30 minutes, the solutions were mixed together for an hour with continuous stirring at 60°C. The solution was then deposited on Si-substrate with optimized layer of thickness. The films were spun onto the wafer with 3000 rpm for 1 minute by using spin coater. The pyrolysis was happened between the layers of films for 2 minutes. The films were annealed later on at 650°C for 10 minutes by using rapid thermal processing (RTP) furnace. The crystallization phase of Sm-doped KNN was analyzed by using X-ray diffractometer (XRD). Then, the electrical properties were measured using LCR meter.

3. RESULTS AND DISCUSSION

The XRD patterns of Sm-doped KNN with different concentrations of 0 up to 0.9 mol % Sm are shown in Fig. 1. It was found that the diffraction peak (200) of KNN has been shifted with the addition of dopant element. Moreover, the addition of Sm has changed the structure of KNN from orthorhombic into tetragonal, thus reducing the lattice distortion [6]. As reported in other work, higher piezoelectric properties might be obtained, however, it depends on the ceramic compositions [7].

The LCR meter have been used to measure the tangent loss (tanδ, %) and dielectric constant (ε'), as shown in Fig. 2. The film with above 0.3 mol % of Sm concentration was found to have better electrical properties. It can be clearly seen that the decrement of tan δ and increment of ε' is attributed due to doping effect. Furthermore, it also indicates that the effectiveness of rare-earth in KNN for better electrical properties in piezoelectric applications.

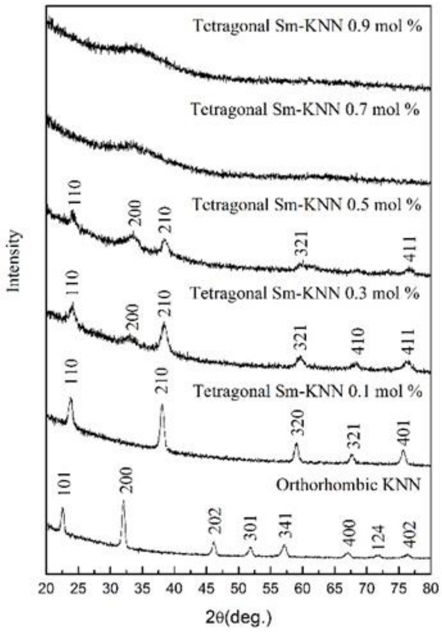

Figure 1 XRD patterns of the KNN thin films with different Sm concentrations of 0, 0.1, 0.3, 0.5, 0.7, and 0.9 mol % deposited on Si substrate and subsequently, annealed at 650°C.

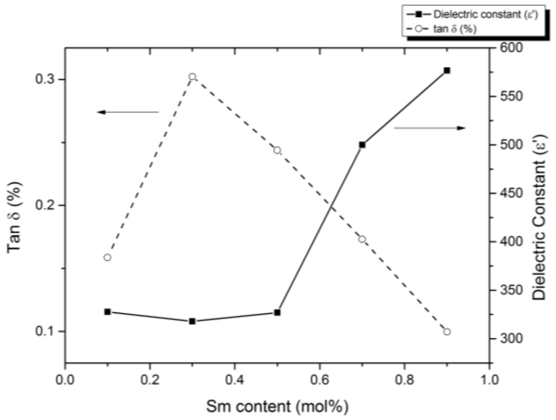

Figure 2 Dielectric constant and tangent loss of KNN thin films with different Sm concentrations.

4. CONCLUSIONS

In this study, the KNN thin films with different Sm concentrations were fabricated to improve its electrical properties. The structure of KNN with higher Sm concentration was found to be transformed from orthorhombic to tetragonal phase, which is also resulted in better electrical properties. This result is believed can contribute in the development of piezoelectric thin films for sensor applications.

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