Investigation of dielectric properties in tantalum doped AgNbO₃ ceramic

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Abstract. Dielectric materials developed from Tantalum (Ta) doped AgNbO₃ (ATN) show excellent properties in variety of electronic technologies. In the temperature range of 70 to 400 °C, four major dielectric abnormalities were observed in ATN (x = 0.1) while in ATN (x = 0.2) the maxima of M₁-M₂ phase shift to low temperature value. ATN ceramics’ dielectric properties dominate temperature and electric field-based performance, which has a major effect on their properties. This study looked into the dielectric properties in ATN.

Keywords: Ag(Nb,Ta)O₃; AgNbO₃; Dielectric properties;

1. Introduction

Researchers are increasingly attracted to lead-free material investigation due to environmental concerns. Among them, lead-free silver based perovskite attract more attention due to their effective properties and performance. In the wide range of silver based perovskite Ag(Nb,Ta)O₃ (ATN) ceramic show excellent dielectric / ferroelectric properties and complex Phase transitions, which have extensive electronic applications such as microwave devices, energy storage devices, wireless-communications techniques, micro-electronics technologies, acoustic wave devices etc [1]. The dielectric properties and phase transition of ATN was first inspect by kania [2]. Later Pawelczyk describe structural information based on XRD pattern to determine phase transition of ATN, were at increasing temperature a series of phase transition M(M₁, M2, M3), O(O₁,O2), T and C was described. Among these different phases M phases shows unique properties near room temperature in ATN ceramics for x < 0.9 composition [3]. Ph Sciau et al. identified centrosymmetric space group (Pbcm) which signified antiferroelectricity for all three M phases [4]. Later Yashina et al. redefine the non-centrosymmetric space group (Pmc2₁) for phase M₁, which determine ferroelectricity in parent AgNbO₃ which contradict early assigned centrosymmetric Pbcm space group [5]. At x = 0.5, Valant et al. observed a high permittivity (εr > 400) in ATN that can be used in the radio and microwave frequency ranges [6]. In ATN the physical properties like dielectric losses, relative permittivity, ferroelectric response depends on processing parameters’ as well as Nb/Ta ratio [7-9]. As a result, it is important to well-ordered study the ATN solid solution. The dielectric properties of ATN for x = 10 and 20 % molecular weight of Ta₂O₅ were investigated in this study. The aim of the alterations is to introduce the impact of minute insertion of b-site dopant in crystal structure and analyze the possible impacts on dielectric properties.

2. Experimental Procedures

The solid-state method was applied to produce the samples of Ta doped AgNbO₃ (ATN) ceramics from powdered silver oxide Ag₂O (99.5%), niobium pentaoxide Nb₂O₅ (99.99%) and tantalum
pentaoxide Ta₂O₅ (99.99%). Materials were dried for 2 hrs at 200 °C to remove moist before being weighted in stoichiometric proportions. In the first stage, Nb₂O₅ and Ta₂O₅ and manually mixed for 4 hrs, which are reacted for 20 hrs at 1200 °C to obtain sample homogeneity and equilibrium. Following that, Ag₂O was added to the first-stage mixture and blended for 4 hrs, after that obtain mixture was calcined for 10 hrs at a temperature of 1050 °C. The calcined samples were mixed again, compacted into discs and sintered at 1150 °C for 10 hrs. The phase characteristic and crystallinity of prepared samples are described by XRD, while the surface topography is shown by SEM. The grain sizes and crystallites of the samples are defined by SEM and XRD data. In different frequency ranges, the deviation in loss tangent (tan δ) and dielectric permittivity (ε) was calculated.

3. Result and Discussion

3.1. Structural Analysis

Figure 1 depicts the XRD graph of the prepared ceramic. All of the XRD peaks correspond to orthorhombic AgNbO₃ and are well indexed, confirming the formation of necessary phases [10-13]. The location of all the diffraction peaks in the doped system matches that of the parent one. The relocation of the XRD pattern may be due to the sintering result or it could be due to the partial replacement of Ta⁵⁺ in site of Nb⁵⁺ in the perovskite structure. SEM image in Figure 2 define smaller grain size with a variance range of 2 – 3 µm as compared to parent AgNbO₃. The holes became wider (2 – 3 µm) and integrated because tantalum shows refractory behavior and high sintering temperature also required for the mixture formation, but due to this higher sintering temperature of the mixture, silver oxide (Ag₂O) decompose and white spots are clearly visible on the surface of mixture. Ta (Tantalum) inclusion in spite of Nb (Niobium) results in no discernible change in the form of polyhedron grains in the AgNbO₃ system.

![Figure 1. The XRD plot of ATN ceramics at room temperature.](image)
3.2. Dielectric and Ferroelectric study

The dielectric dispersion and loss tangent measurement of ATN ceramic samples are carried on frequencies ranging from low 1 kHz to high 1 MHz. Figure 3 and Figure 4 shows dielectric dispersion and loss tangent as a function of temperature noted for 1 MHz. Four major dielectric abnormalities at 85 °C (M₁-M₂), 270 °C (M₂-M₃) and 360 °C (M₃-O₁), are noted in ATN (x = 0.1) ceramic sample, which almost match previous records: phase M₁ (ferroelectric), phases M₂ - M₃ (antiferroelectric) and phases O₁ - O₂ (paraelectric) [14-17]. For ATN (x = 0.2), the plot similarly resemble with parent AgNbO₃ but the maxima of M₁-M₂ switch to low temperature value as compare to AgNbO₃ while M₂-M₃ and M₃-O₁ value is 165 °C and 355 °C, respectively. In addition a minor peak appears in the M₂ region at around 70 °C which is also seen in AgNbO₃.

Figure 2. The SEM pattern of (a) ATN (x = 0.2) and (b) ATN (x = 0.1) ceramics at room temperature.

Figure 3. Dielectric dispersion of (a) ATN (x = 0.1) and (b) ATN (x = 0.2) ceramics at 1 MHz frequency.
However, the ferroelectric transition temperature recorded 67 °C to 70 °C, appears to be a little higher in ATN (x = 0.1). Previous research support that the concentrations of Ag^{1+} ions affect the fluctuation of the ferroelectric transition temperature peak [18]. The graphs show the dielectric permittivity (\epsilon) of ATN match with AgNbO_3 system were the Ta (Tantalum) replacement for Nb (Niobium) has resulted in a lower dielectric loss value.

4. Conclusion

The standard solid state technique was used to prepare the Ta modified ceramic sample of AgNbO_3 and characterized using XRD and SEM for ceramic phase and surface morphology analysis. For uniform distribution of Ta and Nb a higher sintering temperature is required which also decompose the ATN perovskite phase. Four major dielectric abnormalities were remarked in 70°C to 400 °C in ATN (x = 0.1) while in ATN (x = 0.2) the M_1-M_2 phase shift to low temperature value.

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