



The Electrical Characterization and Relaxation Behavior of $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ Ceramics

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$\text{Ag}(\text{Ta},\text{Nb})\text{O}_3$ materials have a perovskite structure with a low loss tangent. These materials have been widely researched for their applications as high-frequency, passive components. Also, $\text{Ag}(\text{Ta},\text{Nb})\text{O}_3$ materials have weak frequency dispersion with high dielectric permittivity which gives them enormous potential for use in electronic components, including the filters, and embedded capacitors. Therefore, our research will discuss the structural and electrical relaxation properties of $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics for device applications. We will investigate using X-ray diffraction to understand their structural properties and will analyze voltage dependent leakage current and time-dependent relaxation behavior to understand their material properties.

Keywords: $\text{Ag}(\text{Ta},\text{Nb})\text{O}_3$ ceramics, XRD, Relaxation current

1. INTRODUCTION

These days, perovskite-type ferroelectric materials have drawn substantial attention due to their properties for dielectric and piezoelectric applications, like for capacitors or infrared sensors [1]. In this paper, we introduce $\text{Ag}(\text{Ta},\text{Nb})\text{O}_3$ materials which have highly dielectric properties and low dielectric losses. These materials are potentially useful for wireless-communications, microelectronic technologies, and the miniaturization of micro-

wave components [2-4]. Comprehensive study of the dielectric properties of microwave, and sub-millimeter to infrared spectroscopy have proved that $\text{Ag}(\text{Ta},\text{Nb})\text{O}_3$ are good ferroelectric materials [5].

Ferroelectrics are brilliant materials which have a nonlinear behavior in their dielectric permittivity, with an applied electric field and ferroelectric characteristics strongly dependent on composition, processing condition, microstructure, applied electric field, operating temperature, and frequency. $\text{Ag}(\text{Ta},\text{Nb})\text{O}_3$ (hereafter ATN) material can be adapted to microwave applications due to their low loss tangent of around 3×10^{-3} from the 1 kHz to the 100 GHz range [6]. Many studies have been carried out to investigate how to improve the dielectric properties through a conventional mixed method [7-10]. ATN shows an interesting dependence of physical properties, such as phase transition and

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the dielectric permittivity on the Ta/Nb com-position. According to Kania, perovskite ATN ceramics showed high dielectric permittivity and low loss tangent with temperature-stable dielectric properties [6]. Therefore, ATN can be a useful material for applications in wireless communications, microwave technologies, and miniaturization of components [11]. The electrical properties, including the leakage current and the relaxation behavior, were found to be greatly influenced by the composition. From this point view, we analyze the characteristics of the various compositions of ATN ceramics.

2. EXPERIMENTS

To prepare Ag(Ta,Nb)O₃ ceramics, Ag(Ta,Nb)O₃ powder was prepared from Ag₂O, Ta₂O₅, and Nb₂O₅ powders with a high purity of 99.9 % by a conventional mixing method. Ag₂O, Ta₂O₅, and Nb₂O₅ powders were employed to prepare the stoichiometric composition of Ag(Ta,Nb)O₃ ceramics. After being ball-milled for 24 hours with ZrO₂ balls and then dried, Ag(Ta,Nb)O₃ powders were calcined at 940°C for 2 hours and then slowly cooled. According to the different Ta / Nb compositions of Ag(Ta,Nb)O₃ materials, the sintering temperatures have to be different [12]. The refined powder size was of around 127 μm. After granulation, these powders were pressed at 1 ton/cm². After pressing, the Ag(Ta,Nb)O₃ ceramics were sintered at 1,140°C for 4 hours.

Finally, silver paste was applied to the upper and bottom side of the Ag(Ta,Nb)O₃ ceramics. We measured the crystalline structure of the Ag(Ta,Nb)O₃ ceramics using X-ray diffraction (XRD) patterns (θ-2θ scans with Cu-Kα source). The time-dependent leakage current behaviors of the ATN ceramics were measured using a Keithley 6517A Electrometer/High Resistance Meter.

3. RESULT AND DISCUSSION

Figure 1 displays the θ-2θ X-ray diffraction patterns of Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics, which were sintered at 1,140°C. According to the analysis of the X-ray diffraction patterns, we found that Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics have a perovskite structure and don't observe the pyrochlore phase. The lattice parameter *c* and *a* of Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics were calculated by employing the Nelson-Riley extrapolation function with a least mean squares method:

$$\frac{C_{\cos\theta} - C_0}{C_0} = A \cdot \cos^2\theta \left(\frac{1}{\sin\theta} + \frac{1}{\theta} \right) \quad (1)$$

where *C_{cosθ}* is an interplane distance calculated from the apparent Bragg 2θ peak positions, and *A* is a fitting coefficient. The calculated lattice parameters *c* and *a* of the Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics were 3.957 Å and 3.937 Å. According to these lattice parameters, we suppose that these ATN ceramics have perovskite structure with a pseudo-cubic system [12].

Figure 2 illustrates the temperature dependent resistivity of the Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics. The resistivity of these ceramics was calculated from the resistance by considering the geometry. During calculation, the total electrode area of the vertical-type ceramic capacitor and the length of the current path between the electrodes were considered. Due to the current-voltage properties and their relationship to temperature, we proposed the Arrhenius equation to describe the temperature-dependent resistance behaviour:

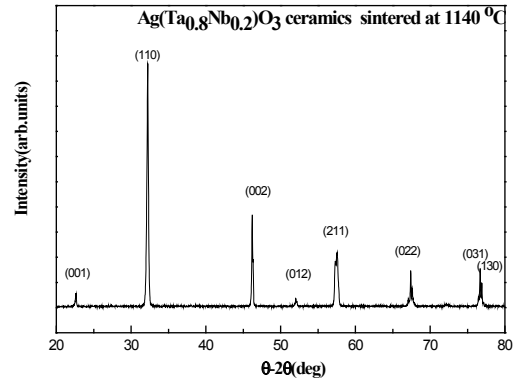


Fig. 1. X-ray diffraction (XRD) θ-2θ scans with CuKα radiation for Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics.

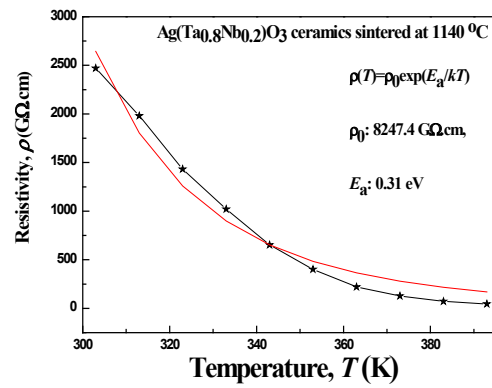


Fig. 2. Temperature-dependent resistivity of the Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics in the Arrhenius form.

$$\rho(T) = \rho_0 \exp\left(\frac{E_a}{kT}\right) \quad (2)$$

where *T* is the temperature in Kelvin, *E_a* is the activation energy of carriers, *k* is the Boltzmann constant (8.62×10⁻⁵ eV/K), and *ρ₀* is the resistance at infinite temperature. The temperature coefficient of resistance, *α*, can be written as

$$\alpha = \frac{1}{\rho} \frac{d\rho}{dT} = \frac{E_a}{kT^2} \quad (3)$$

From Eq. (3), the calculated temperature coefficient of this thick film was -5.08×10⁻¹¹ K⁻¹. The simulated *ρ₀* of the Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics was 8,247.4 GΩ.cm, and the calculated activation energies were around 0.31 eV [13].

Figure 3 indicates the transient current *I* recorded in the time domain for the Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics. At a room temperature range, we measured the time-dependent leakage currents at voltages of 30, 60, and 90 V, respectively. By applying a respective step voltage to the Ag(Ta_{0.8}Nb_{0.2})O₃ ceramics, we were able to presume that the bias voltages caused different types of dipole reorientations in the ceramics.

Therefore, we defined these time-dependent leakage currents as polarization currents 'I_{transient}' from the different types of dipole motions and the true leakage current 'I_{true}'. From the simulation in Fig. 3, we may claim that the time-dependent current behavior can be expressed by the relaxation of dipole motions in the

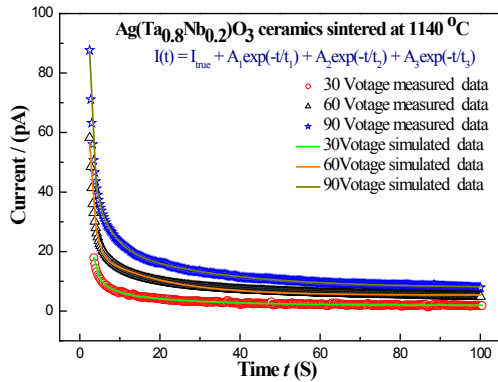


Fig. 3. Time-dependent leakage current characteristics of $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics.

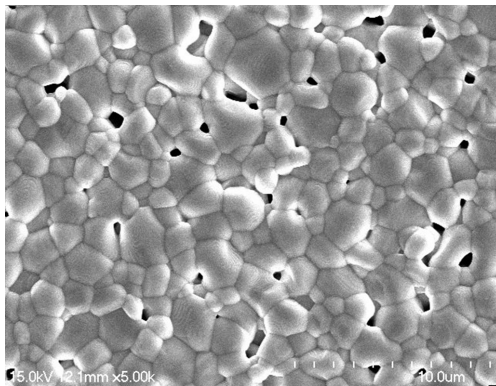


Fig. 4. The SEM image of the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics sintered at $1,140^\circ\text{C}$.

polarization process [14]:

$$I(t) = I_{true} + A_1 \exp(-t/t_1) + A_2 \exp(-t/t_2) + A_3 \exp(-t/t_3) \quad (4)$$

The simulated values are listed in Table 1.

Figure 4 shows the SEM image of the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics sintered at $1,140^\circ\text{C}$. We observed the grain shape and size of the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics, and we can argue that the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics were well crystallized. Due to the volatility of Ag ions during the sintering process, a small porosity of the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics was observed. We can conclude that the porosity and grain size in the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics played a role in increasing the transient current I recorded in the time domain for the $\text{Ag}(\text{Ta},\text{Nb})\text{O}_3$ ceramics.

4. CONCLUSIONS

In this research, we analyze the structural and electrical characteristics of the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics. Using X-ray diffraction analysis, we could see that ATN ceramics had a perovskite structure with pseudo-cubic systems. The calculated lattice

Table 1. Fitting value of the transient current I recorded in the time domain for $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics.

	$\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics		
	30 V	60 V	90 V
I_{true}	1.8972×10^{-12}	5.3454×10^{-12}	7.7909×10^{-12}
A1	1.4313×10^{-11}	5.3496×10^{-11}	1.8951×10^{-9}
t1	3.8391	1.1321	0.641
A2	5.4443×10^{-12}	1.5369×10^{-11}	4.8672×10^{-11}
t2	21.6673	16.5071	3.8471
A3	5.7002×10^{-10}	4.2877×10^{-13}	1.7008×10^{-11}
t3	0.7601	16.5681	26.4724

parameters c and a of the $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics were 3.957 \AA and 3.937 \AA . Using the temperature-dependent resistivity, we calculated the temperature coefficient of this thick film to be $-5.08 \times 10^{-11} \text{ K}^{-1}$. The simulated ρ_0 was measured at $8,247.4 \text{ G}\Omega\cdot\text{cm}$, and the calculated activation energies were of around 0.31 eV . Also, according to the analysis of the time-dependent relaxation behaviors, we may argue that the time-dependent relaxation behaviors of the ATN ceramic's different types of dipole reorientations were caused by range of the bias voltage.

We conclude $\text{Ag}(\text{Ta}_{0.8}\text{Nb}_{0.2})\text{O}_3$ ceramics had a well crystallized structure and brilliant electrical properties for application as passive components.

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