

Dielectric Properties of Eco-Friendly Silver Sodium Niobate Perovskite Ceramic

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ABSTRACT

Polycrystalline sample of Silver Sodium Niobate $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ (ANN) had been fabricated by solid state reaction method for its systematic dielectric investigation. X-ray diffraction (XRD) had been used to get information regarding the structure of the fabricated sample and the specimen was found to exhibit perovskite orthorhombic structure at room temperature. To analyse microstructure of the specimen, Scanning Electron Microscope (SEM) & Energy Dispersive X-Ray Spectroscopy (EDX) had been used. Dielectric constant and tangent loss of the synthesized ceramic had been measured at various frequencies ranging from 10 Hz to 10^6 Hz corresponding to different temperatures between 25 °C to 350 °C. Both dielectric parameters had been found to be decreasing with increase in frequency at all temperatures.

Keywords: structural, perovskite, dielectric constant, tangent loss

INTRODUCTION

Perovskite oxides form an interesting class of materials as they possess unique properties suitable for many different applications. A perovskite oxide is represented by general formula ABO_3 . In ABO_3 , A represents a bigger cation generally monovalent or divalent belonging to alkali, alkaline or a rare earth family, while B represents a smaller cation usually tetravalent or pentavalent belonging to transition metal. [1-3] They have stable structure. So, it is possible to replace cation present at A and/or B -site by many different cations leaving parent structure undisturbed.[4] This substitution leads to enhancement of their dielectric properties. These ABO_3 ceramics are used in a wide variety of electronic devices like capacitors, sensors memory devices etc. These applications require mostly lead based materials. But use of lead-based materials had been restricted worldwide due to its toxic nature. In search of lead-free materials, a lot of research work had been done. Structural investigation of La-modified silver niobate ceramic $\text{Ag}_{1-x}\text{La}_x\text{NbO}_3$ (with $x=0.005$) had been done.[5] Structural, FTIR and ac conductivity of NaMeO_3 ceramics (Me= Nb, Ta) had been studied.[6] Study of dependence of dielectric properties on temperature of sodium potassium niobate ceramics $\text{Na}_{0.5}\text{K}_{0.5}\text{NbO}_3$ had been performed.[7] Y_2O_3 ceramics with Nd^{3+} and Er^{3+} dopants and $\text{ZrO}_2/\text{La}_2\text{O}_3$ sintering additives had been synthesized using co-precipitation method for study of structural and optoelectrical properties.[8] BCZT ceramics with Sm^{3+} and Fe^{3+} dopants had been explored for understanding their electrocaloric properties. [9] Present work is to synthesize and explore the dielectric properties of lead-free eco-friendly silver sodium niobate ceramic with composition $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$.

EXPERIMENTAL PROCEDURE

Ceramic sample of silver sodium niobate with composition $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ had been synthesized by solid state reaction method. Highly pure starting raw materials used were silver oxide (Ag_2O), sodium carbonate (Na_2CO_3) and niobium pentaoxide (Nb_2O_5). After mixing and grinding these powders, calcination had been performed at 1050° for 2 hours. Adding few drops of polyvinyl alcohol (PVA), pellets were formed with 10 mm diameter and 1 mm thickness by hydraulic press applying 1 ton pressure. Thereafter, sintering had been performed at

temperature 1150° C for 1 hour duration. Structural investigation of powdered ceramic sample had been done using X-ray diffraction (XRD) corresponding to room temperature using Phillips X-ray diffractometer with Cu K α radiations in 2 θ ranging from 20° to 80°. Microstructural analysis had been carried out using Scanning Electron Microscope (SEM) (TESCAN VEGA III LM) along with EDX. For measurement of dielectric properties, sintered pellets had been converted into capacitor configuration by applying thin paste of silver on it's both sides. Dielectric measurement had been carried out by Dielectric/ Impedance analyzer (Keysight Impedance analyzer E 4990 20 Hz- 20 MHz).

RESULTS AND DISCUSSION

Structural Analysis- Recorded XRD pattern of ANN specimen is revealed in Figure 1. Joint Committee on Powder Diffraction Standards data card n. 01-082-0606 (NaNbO $_3$) had been used for indexing of peaks.[10] XRD data analysis confirmed the presence of stable perovskite orthorhombic structure of the specimen.

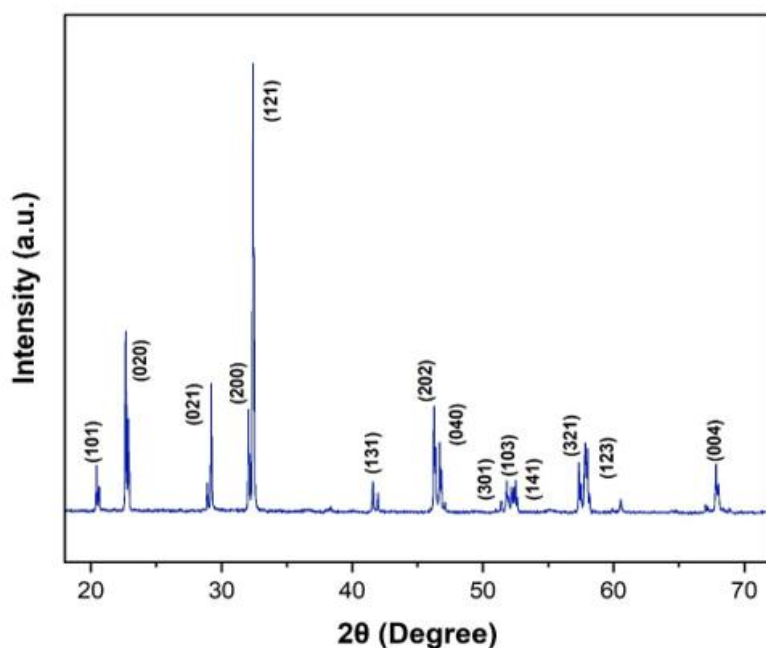
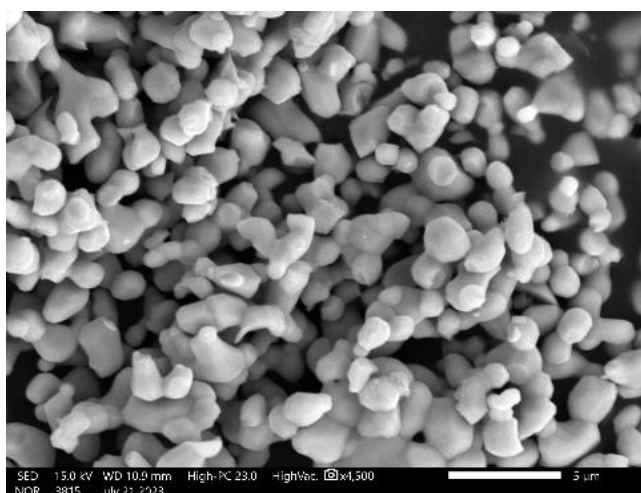


Figure 1: XRD pattern of Ag $_{0.2}$ Na $_{0.8}$ NbO $_3$

Microstructural Analysis- Micrograph of specimen obtained by SEM has been shown in Figure 2. Figure 2 indicated the presence of grains with size different from each other and average grain size of 1.69 μ m had been calculated using ImageJ software. Recorded EDX spectrum confirmed presence of all required elements in sample.

a)



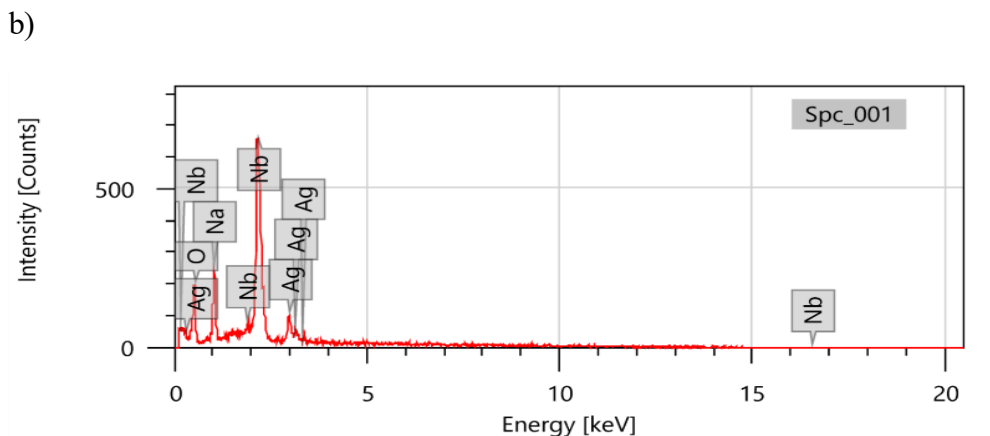


Figure 2: (a) SEM image, (b) EDX spectrum of $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$

Dielectric Analysis

In order to understand dielectric properties of synthesized ANN specimen, variation of dielectric constant and tangent loss i.e. $\tan \delta$ against frequency corresponding to room temperature has been plotted in Figure 3 and Figure 4 respectively.

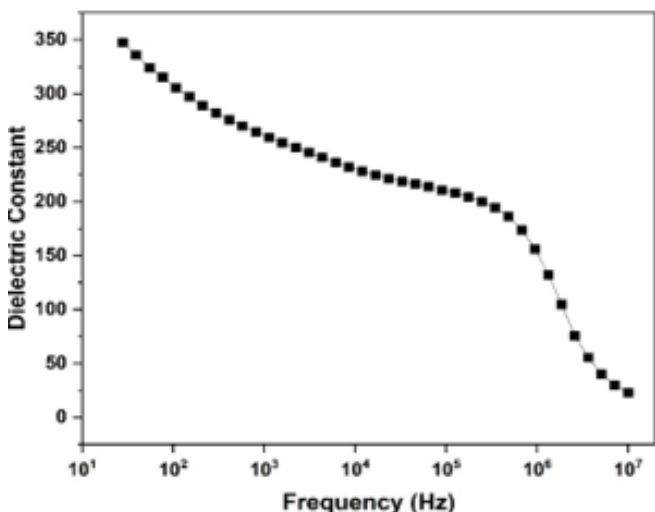


Figure 3: Variation of dielectric constant with frequency for $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ at room temperature

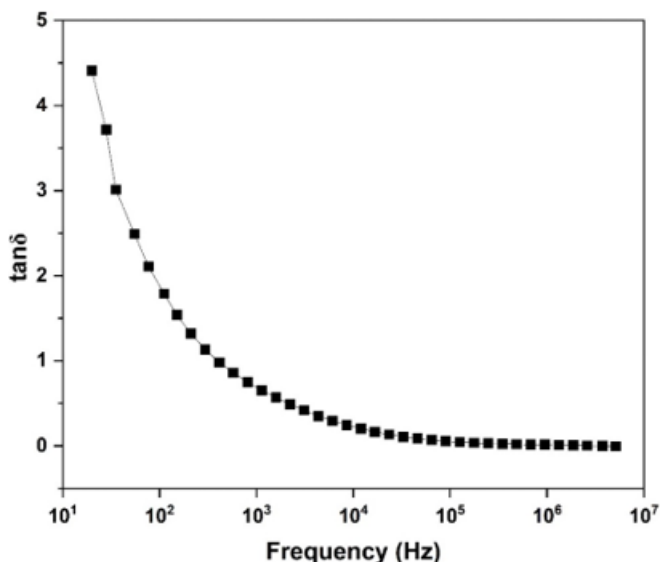


Figure 4: Variation of $\tan \delta$ with frequency for $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ at room temperature

Figure 3 demonstrated that dielectric constant is large at low values of frequency. But as frequency goes on increasing, a reduction in dielectric constant was revealed. This decrease in dielectric constant with frequency is known as dielectric dispersion.[11] This nature of behaviour of dielectric constant had also been already reported.[12] This variation can be explained in terms of Maxwell-Wagner model for space charge polarization.[13] This model considers the dielectric material to be made up of grains having large conductivity separated by grain limits possessing low conductivity. At the small frequencies, the charge carriers get sufficient time to reach grain limits by moving between grains thus developing large interfacial polarization which in turn leads to high dielectric constant. As frequency increases, there are less chances of charge carrier to reach at grain boundary leading to less polarization and low dielectric constant.[14] $\tan \delta$ also behaves in similar manner showing reducing trend with increasing frequency as shown in figure 4. A high value of $\tan \delta$ has been revealed by figure 4 in this sample at low frequencies. This is due to conduction loss because of an increase in a.c conductivity. It happens as a result of increase in hopping rate of charge carriers thus giving rise to their long-range migration at low frequency. At high frequencies, it is almost constant. This is because with rise in frequency of applied field, space-charge, orientational and ionic polarization lag behind field causing a decrease in tangent loss. Figure 5 and Figure 6 display the evolution of dielectric constant & $\tan \delta$ of sample with the frequency at various values of temperature.

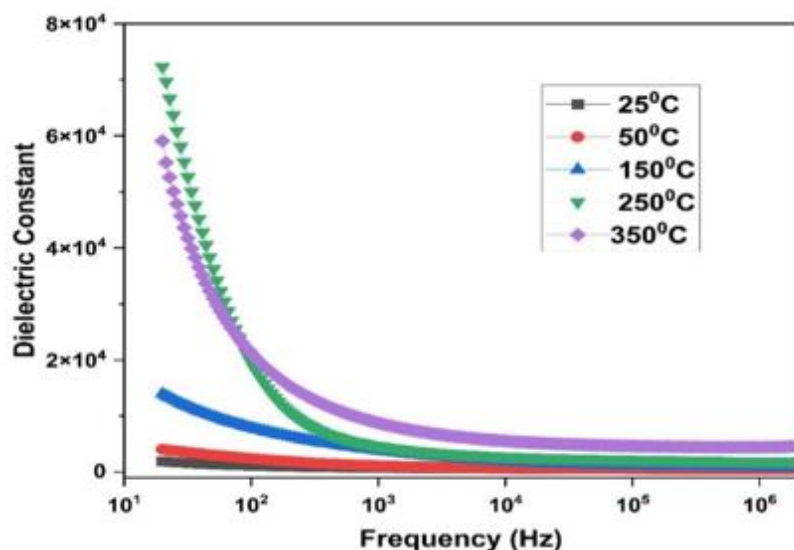


Figure 5: Variation of dielectric constant with frequency for $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ at various temperatures

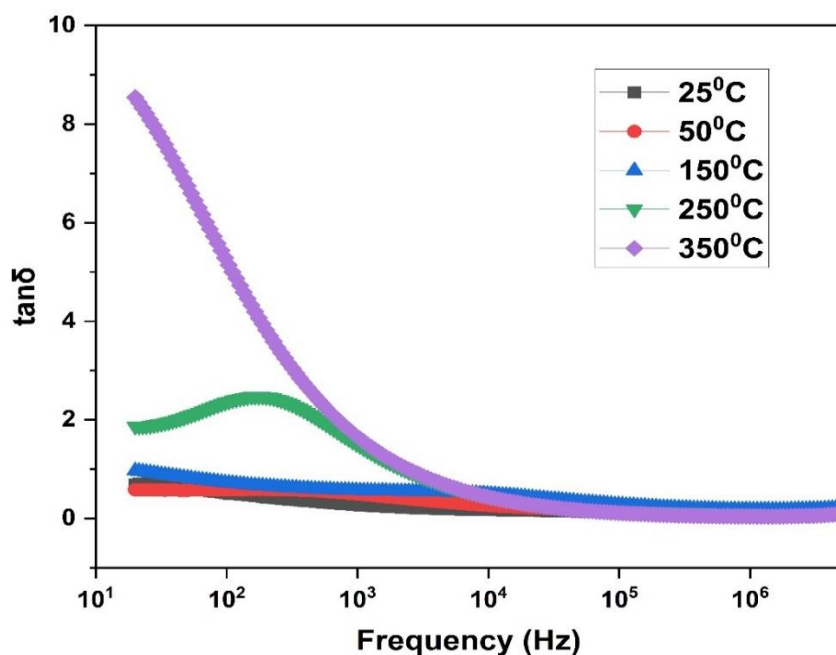


Figure 6: Variation of $\tan \delta$ with frequency for $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ at various temperatures

It has been observed from figure 5 and figure 6 that at any frequency, with rise in temperature, both the dielectric constant and $\tan \delta$ rise. This can be explained by the fact that due to rise in temperature, mobility of charge carriers increases thus enhancing space-charge polarization and hence dielectric constant. But this also leads to weakening of process of dielectric relaxation. Therefore, more energy will be dissipated leading to high tangent loss. High $\tan \delta$ at elevated temperatures is found to be due to accumulation of charges at grain boundaries. Another factor for rise in $\tan \delta$ is that alignment of molecular dipoles will also get affected due to increased thermal agitation at high temperatures. Values of dielectric constant & $\tan \delta$ corresponding to various frequencies at different temperatures have been put respectively in Table 1 & Table 2.

Table 1: Values of dielectric constant for $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ corresponding to various frequencies at different temperatures

Frequency	Temperature				
	25°C	50°C	150°C	250°C	350°C
20 Hz	1924.11	4054.12	13938.23	72286.57	59098.01
100 Hz	1164.99	2393.80	7802.50	18830.88	20837.50
500 Hz	840.71	1381.75	4753.65	6083.75	10616.48
1 KHz	759.61	1126.43	3808.28	4564.58	8672.19
5 KHz	638.64	818.34	2309.53	3081.83	6119.37
10 KHz	598.45	745.97	1855.12	2752.93	5525.83
50 KHz	524.16	623.71	1297.33	2350.72	4810.00
100 KHz	501.01	583.14	1169.37	2227.96	4638.72
500 KHz	466.35	521.11	964.69	2004.05	4457.84
1000 KHz	454.84	503.86	905.85	1940.32	4454.76

Table 2: Values of tangent loss ($\tan \delta$) for $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ corresponding to various frequencies at different temperatures

Frequency	Temperature				
	25°C	50°C	150°C	250°C	350°C
20 Hz	0.6907	0.5813	0.9710	1.8630	8.5437
100 Hz	0.5017	0.5811	0.7212	2.4145	5.1418
500 Hz	0.3281	0.5552	0.5943	2.0080	2.3952
1 KHz	0.2648	0.4888	0.5655	1.4667	1.6174
5 KHz	0.1824	0.3189	0.5339	0.6388	0.6489
10 KHz	0.1662	0.2577	0.4927	0.4273	0.4198
50 KHz	0.1322	0.1772	0.3396	0.1949	0.1647
100 KHz	0.1147	0.1525	0.2759	0.1519	0.1073
500 KHz	0.0836	0.1055	0.1911	0.1058	0.0482
1000 KHz	0.0829	0.0955	0.1779	0.0928	0.0408

High value of dielectric constant marks the utility of this ANN specimen in applications like multilayer capacitors and memory devices.

CONCLUSION

Perovskite silver sodium niobate $\text{Ag}_{0.2}\text{Na}_{0.8}\text{NbO}_3$ had been formed by solid state reaction. Structure of specimen had been found to be perovskite orthorhombic crystal structure with an average grain size of 1.69 μm . At a particular temperature, a reducing nature of dielectric constant & $\tan \delta$ with rise in frequency had been depicted. But an increase in both of these parameters had been observed as temperature increased. Obtained values of

dielectric constant signifies this ANN specimen suitable for making the multilayer capacitors and memory devices.

Conflict of interest- The authors declare that they have no conflict of interest.

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