

The effect of silica addition on dielectric properties of barium titanate nanoparticles

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Abstract

The effect of silicon dioxide (SiO_2) nanoparticles on the dielectric constant was studied. Different weight percentages of silica (3, 5, and 7 %) were added to BaTiO_3 nanoparticles. The structure, morphology, and dielectric properties were obtained using X-ray diffraction (XRD) and scanning electron microscopy (SEM), and an LCR meter. The diffraction patterns were shown that the crystallite size of BaTiO_3 increased with the addition of SiO_2 . The dielectric constant and the dielectric loss were measured with various temperatures ranging from 27 to 180 °C at a constant frequency of 10 kHz. It was found that the value of the dielectric constant for the pure BaTiO_3 is the highest at the Curie temperature (128 °C). The Curie temperature shifted to a higher value and the dielectric constant was decreased when the silica was added to BaTiO_3 . Moreover, with increasing the silica percentage, the variation in the dielectric loss values with the temperature was observed.

Keywords: Barium titanate, silicon dioxide, dielectric constant, Curie temperature, dielectric loss.

1. Introduction

Nanoparticles are substances having unique nanometer-scale characteristics and structures. Because of their small size, significant surface area-to-volume ratio, and quantum effects, these materials have distinct physical, chemical, mechanical, and optical

properties when compared to their bulk materials. Therefore, many research has been done in a variety of scientific areas, including physics, chemistry, materials science, biology, and engineering [1-7]. In the dielectric field, the barium titanate (BaTiO_3) nanoparticles, which are one of the

perovskite materials have been shown to have dielectric properties different than their bulk counterparts [8-10]. BaTiO₃ was studied with the addition of other materials, such as silicon dioxide (SiO₂), and aluminum oxide (Al₂O₃). The stability of this material and the ability to replace the atoms in the crystal lattice with atoms of other substances opened the wide field for many studies based on the development of these materials and improving their structural and electrical properties. It was found that the fusion of a titanium atom with a silica atom in the crystal structure of BaTiO₃ leads to an improvement in the insulating properties of this material and changes the dielectric constant [11-12]. The dielectric constant is also affected under applied temperature. The phase of BaTiO₃ transfers from a tetragonal to cubic at the temperature of (120 – 130 °C), which is a Curie temperature TC [13-14]. The Curie temperature decreases with an increase in silicon dioxide content [16]. Moreover, the dielectric and electrical properties of BaTiO₃/SiO₂ nanocomposite are affected by the nanocomposite's composition, particle size, the interface between the BaTiO₃ and SiO₂ phases, and the sintering. Studies show that the increment of both the sintering degree and the sintering time leads to an increase in the size of the ceramic granules

and an increase in the density of the samples [15-16]. The research aim is to prepare ceramic samples from the BaTiO₃/ SiO₂ in nanoscale particle size in the form of proportions adding (0, 3, 5, and 7 %) of silica, and study their effect on the dielectric properties of BaTiO₃ under applied temperatures.

2. Experimental

2.1 Materials

BaTiO₃ and SiO₂ nanoparticles with sizes of (50-70 nm) and (20 nm), respectively, were procured from Skyspring Nanomaterials, Inc. The powder was blended with a planetary mill after a high amount of SiO₂ nanoparticles (0, 3, 5, and 7 wt %) were introduced in varying amounts. The granules were then compressed into a 9 mm diameter disk and 1 mm thickness. These samples were pressed with a hydraulic press under a pressure of 6 metric tons for 5 minutes. The pellets were sintered at 1100 °C for three hours.

2.2 Characterization and Measurement

X-ray diffraction (XRD) was used to study the structure of all ceramic samples. It was operated at 30 mA, 40 keV, and CuK α

beamline 1.5406 Å within a 2θ angle range of 10-80°. After the completion of the process of polishing and cleaning the samples, the process of coating samples with a conductive silver paste is carried out. The main objective of the silver coating process is to form electrodes on the two surfaces of the samples. Then, the samples were put in an oven at a temperature of 80 °C for 20 min to remove voids in the surface of the sample and to obtain high adhesion to the surface. The dielectric constant and loss tangent were measured at temperatures starting from 27 to 180 °C with step heating (2 °C/min) using an LCR meter. In all measurements, the frequency was fixed at 10 kHz.

3. Results and discussion

3.1 XRD analysis

XRD measurements of the pure barium titanate and the nanocomposites are shown in (figure 1). The diffraction patterns of the BaTiO₃ displayed the strongest peaks at angles of 31.7, 39.0, 45.1, 45.5, 56.3, and 66.0 that are related to 101, 111, 002, 200, 121, and 220 planes of tetragonal phase, respectively. No diffraction peaks caused by the presence of other impurities were detected in pure BaTiO₃ nanoparticles, which means a high degree of purity of the sample. The diffraction diagrams show that by

increasing the proportions of silica nanoparticles to pure barium titanate, the intensity of some diffraction patterns of BaTiO₃ decreases and the lowest peaks disappear, while the intensity peaks of silica increase. The location of some of them changes, which indicates the effecting of the crystal structure of the compound resulting from changes in the dimensions of the unit cell. The emergence of crystalline stresses resulting from the replacement of titanium ions with silicon ions [11-17]. Therefore, the crystallite size of BaTiO₃ increased with the addition of silica. According to the Debye–Scherrer equation, the size was computed to be 23.07 nm for the pure sample. The size was found to increase around 0.7 nm for every 2 % addition of silica.

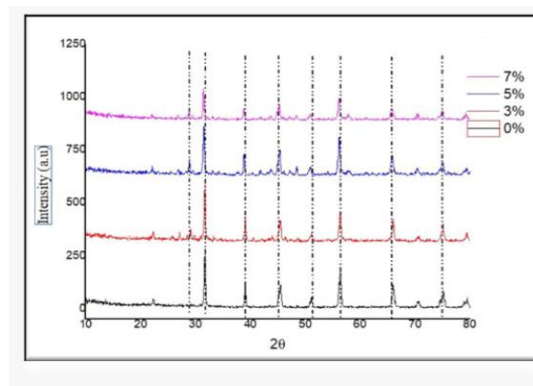


Figure 1: XRD patterns of BaTiO₃ nanoparticles with different amounts of SiO₂: (0, 3, 5, and 7) wt %.

3.2 SEM analysis

SEM images show the surface morphology of both BaTiO_3 and SiO_2 as shown in figures 2, and 2, respectively. In figure 2, the particles of the pure BaTiO_3 sample are shown to be spherical with an average size of 54 nm. In figure 3, the SiO_2 nanoparticles are also displayed in a spherical shape with a diameter of around 12 nm. Moreover, aggregation appeared because of the small particle size of silica.

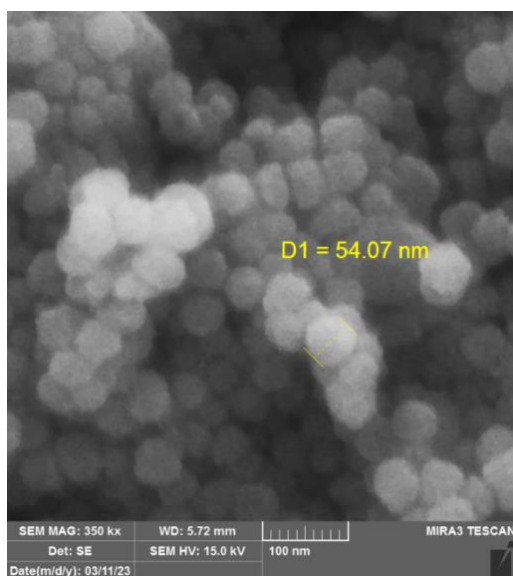


Figure 2: SEM image of BaTiO_3 nanoparticles.

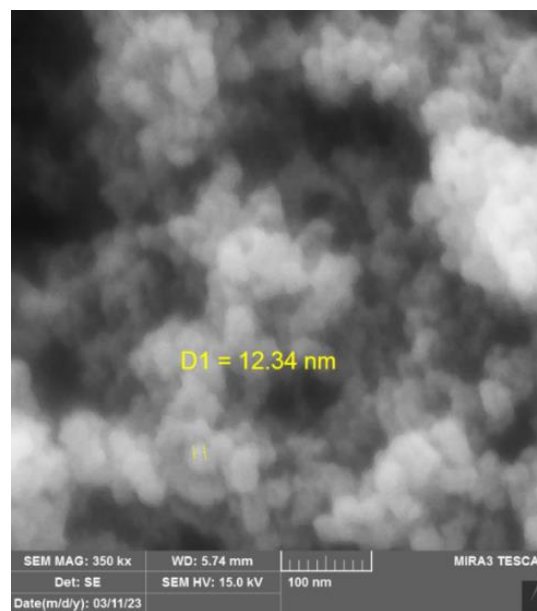


Figure 3: SEM image of SiO_2 nanoparticles.

3.3 Dielectric constant measurement

Figure 4 to 7 shows the dielectric constant of the pure BaTiO_3 and nanocomposites as a function of temperature. The dielectric constant decreased with an increase in the percentage of silicon dioxide because of the decreasing contribution of the electronic polarization. The dielectric constant of all samples increased with increasing temperature until reaching a maximum value at the Curie temperature (128°C), due to more dipoles being drawn toward the electric field as temperatures rise until the resultant polarization approaches its maximum value. The dielectric constant decreases after the Curie temperature due to the disappearance of the electric dipoles. Therefore, the

transition of the material from the tetragonal phase to the cubic phase was done. Moreover, The Curie temperature point shifts to a higher value with increasing the percentage of silica. After cooling, both the Curie temperature degree and the dielectric constant values do not match their values at heating.

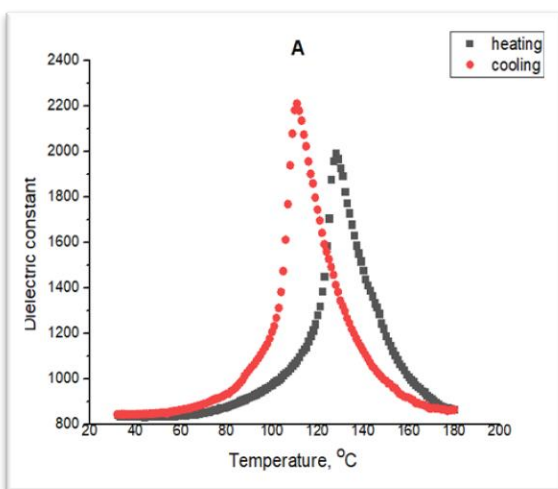


Figure 4: Variation of dielectric constant with temperature at 10 kHz with different weight percentage of SiO₂ is equal to 0 %.

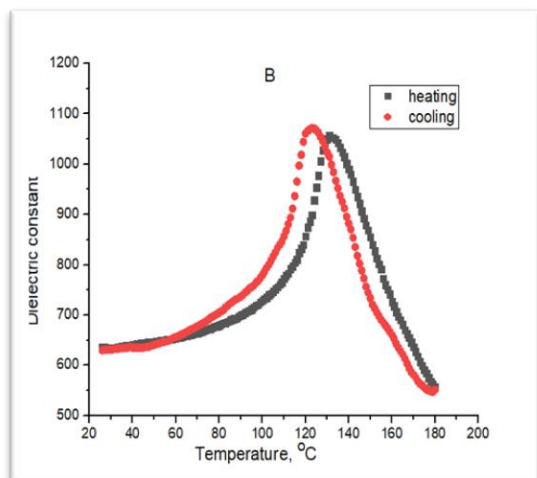


Figure 5: Variation of dielectric constant with temperature at 10 kHz with different weight percentage of SiO₂ is equal to 3 %.

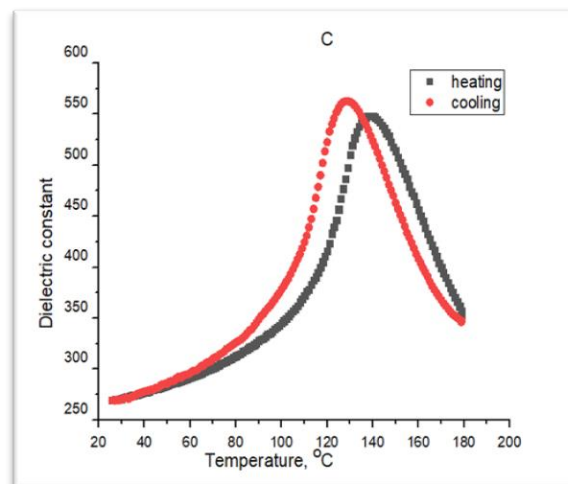


Figure 6: Variation of dielectric constant with temperature at 10 kHz with different weight percentage of SiO₂ is equal to 5 %.

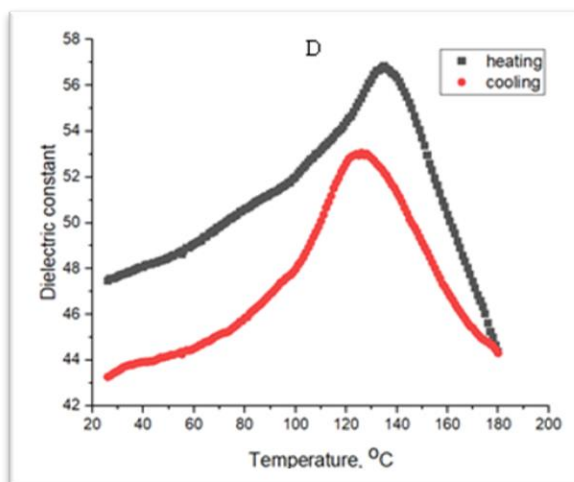


Figure 7: Variation of dielectric constant with temperature at 10 kHz with different weight percentage of SiO₂ is equal to 7 %.

3.4 Dielectric loss measurement

The experimental results showed that the dielectric loss of pure barium titanate was nearly constant before the Curie temperature, while after that it linearly increased with the

temperature as shown in figure 8. However, the dielectric loss of the nanocomposites noticeably decreased with increasing temperatures starting from room temperature to Curie temperature. After the Curie temperature, the values rose. This is because of the phase transformation. Figures 9, 10, and 11 show the dielectric loss of nanocomposites with the addition of SiO_2 . The dielectric loss was more affected by temperature when the percentage of silica was increased.

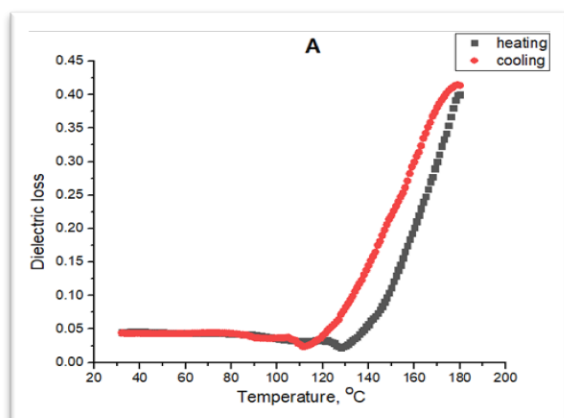


Figure 8: Variation of dielectric loss with temperature at 10 kHz with different weight percentage of SiO_2 is equal to 0 %.

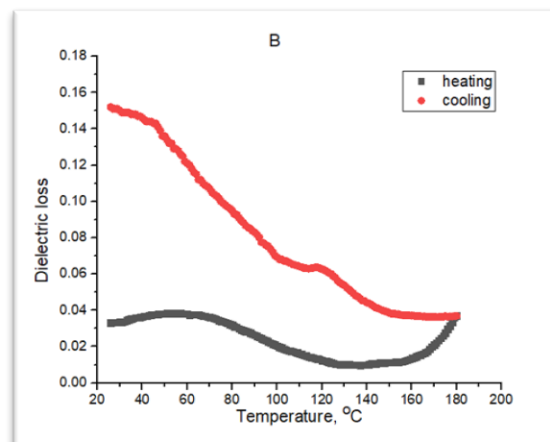


Figure 9: Variation of dielectric loss with temperature at 10 kHz with different weight percentage of SiO_2 is equal to 3 %.

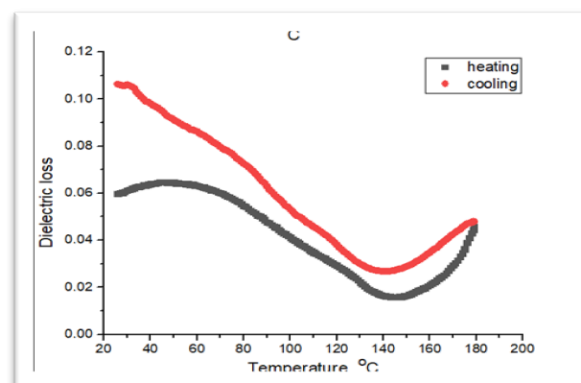


Figure 10: Variation of dielectric loss with temperature at 10 kHz with different weight percentage of SiO_2 is equal to 5 %.

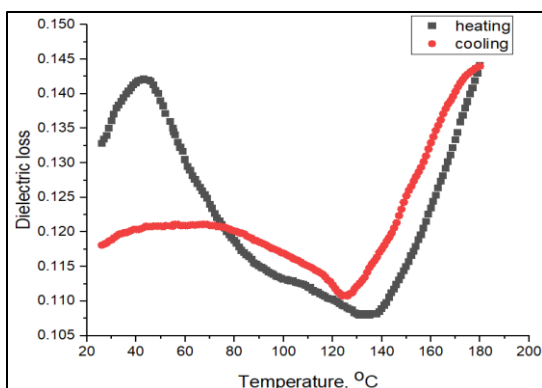


Figure 11: Variation of dielectric loss with temperature at 10 kHz with different weight percentage of SiO₂ is equal to 7 %.

4. Conclusion

The dielectric constant of nanocomposites decreases with increasing the addition of SiO₂ nanoparticles to the pure BaTiO₃ solid solution. As a result of the decreasing contribution of the electronic polarization that depends on the ionic radius. The maximum dielectric constant was shown to be at the Curie temperature for all samples, while the dielectric loss was a lower value at this temperature. The variation in values comes due to the transition from tetragonal toward cubic phase.

5. References

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