RESEARCH ARTICLE

Comparative Study on Electrical and Dielectric Properties of Sintered Nano and Micro Silicon Nitride Ceramics

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ABSTRACT

In the present work we have studied the electrical conductivity, dielectric constant and dielectric loss of Sintered Silicon Nitride ceramics. In this study it was found that the grain size has great impact on electrical conductivity and dielectric properties of Sintered Silicon Nitride Ceramics. The result shows more efficiency of electrical and dielectric properties with nano sized grains. The sintering was performed in a programmable furnace at 950 K. The dc conductivity measured in the temperature range 300 K to 900 K. At higher temperature (T > 800 K), the dc conductivity increases exponentially with temperature for both of the investigated samples. Dielectric constant and loss are measured in the temperature range 300 K to 900 K with frequency range 1 KHz to 1 MHz. To confirm the grain size, the samples are characterized by the Scanning Electron Microscope (SEM). These types of samples can be used as a high temperature semi-conducting material.

1. Introduction

Silicon nitride has attracted considerable interest due to its excellent high-temperature mechanical properties, such as strength, hardness, wear resistance, thermal shock resistance and chemical inertness. This material is utilized for structural applications at high temperatures and its domains of application extend from vessels for chemical reactions to heat-exchanger bearings, engines and gas turbine components [1-3]. Si_3N_4 is also a wide-band-gap semiconductor used in the optical and electrical device industries [4]. In the 20th century, new ceramic materials were developed to use in advanced ceramic engineering; for example, in semiconductors. However, Si_3N_4 nanotubes and nanowires have recently attracted much attention because of their enhanced hardness which is associated with their unique optical and mechanical properties [5-7]. As a result, different physical and chemical routes have been proposed and developed for the synthesis of Si_3N_4 nanowires. Most popular methods for the synthesis of nanowires are chemical vapor deposition [8-9], carbo-thermal reduction of silica and carbon in nitrogen-containing ambient [10-11], nitration of silicon powder [12-13] and the nitration of a Fe–Si catalyst [14]. Silicon nitride ceramics have been frequently used as structural materials for high-temperature applications. They exhibit unique properties because of their high fracture toughness due to a composite microstructure and a high creep resistance controlled by grain boundary. The high-strain-rate plasticity of a silicon nitride ceramic, known as super plasticity, was first reported in 1990 [15]. The report indicated that silicon nitride with a small grain could exhibit plastic behaviour even if it is typically tough and strong. Since 1990, many fine-grained silicon nitride ceramics have been developed.

In this paper we have discussed the electrical and dielectric properties of micro and nano grain sized silicon nitride ceramics for better performance and newer application of the materials.

2. Experimental Details

In this work, we used an α-phase micro powder Silicon Nitride (99.999% from Alfa Aesar) and nano powder (98% from Sigma-Aldrich). Grinding and mixing of micro and nano powder carried out separately in a mortar for 30 minutes to achieve homogeneity. Pellets (diameter~1.00 cm and thickness for Micro~0.08 cm and for Nano~.03cm) of these powders were made using hydraulic press by applying the uniform pressure of 100 MPa, which results in fully isotropic material properties. The micro powder pellets were sintered at 600 ^{0}C for 2 hrs to achieve strength and pellets of nano powder were not sintered to avoid grain growth and agglomeration.
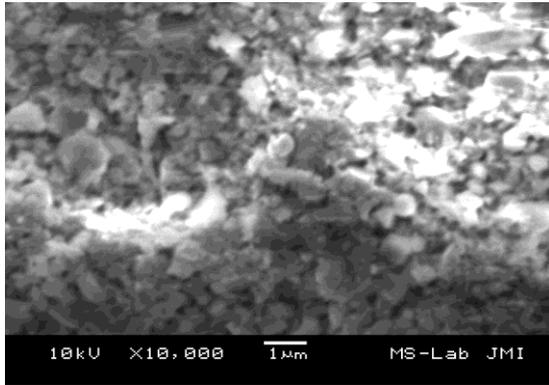
Figure 1(a): *SEM micrograph of micro sized Si$_3$N$_4$ ceramic*
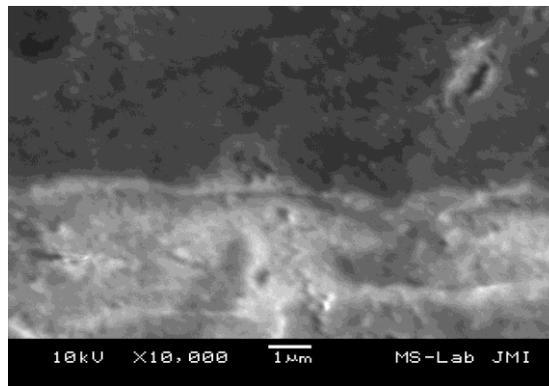
Figure 1 (b): *SEM micrograph of nano sized Si$_3$N$_4$ ceramic*

Scanning Electron Microscope (SEM) has been used for structural characterization. The SEM images of fractured surface for the both, micro and nano sized Si$_3$N$_4$ ceramic samples are shown in the figure 1. A specially designed metallic sample holder is used for DC as well as AC measurements. The sample holder consists of two parts; the upper part contains two steel electrodes passes through Teflon feed, between which the pellet samples were mounted via a screw arrangement. The lower part contains a heating element in the bottom to heat the sample. For both DC and AC measurements, the vacuum of the order 10^{-3} Torr has been maintained inside the sample holder. The temperature was measured by mounting a k-type Chromel-Alumel thermocouple near the sample. A Keithley picoammeter (model 6485) was used to measure the currents which are typically of the order of a few pico-amperes at low temperature and increases on increasing the temperature. For dielectric properties, the capacitance C and dissipation factor D is measured by using Wayne Kerr 4300 LCR meter (frequency range 20 Hz-1 MHz with 0.1% basic accuracy).

3. Results and Discussion

3.1. DC Conductivity

In figure 2, the dc conductivity for both samples has been plotted as a function of reciprocal temperature. Initially, in the lower temperature's region (T<800K), the conductivity is nearly temperature independent for both micro sized as well as nano sized Si$_3$N$_4$ ceramics. In the high temperatures region (T>800 K), the dc conductivity increases exponentially with temperature for both of the investigated samples. Both of the samples shows double slope and the high temperature regions are well fitted to Arrhenius relation-

$$\sigma_{dc} = \sigma_0 \exp(-\frac{\Delta E_{dc}}{kT}) \qquad (1)$$

Where σ_0 is the pre-exponential factor, ΔE$_{dc}$ is the activation energy for dc conductivity and k is Boltzmann constant.
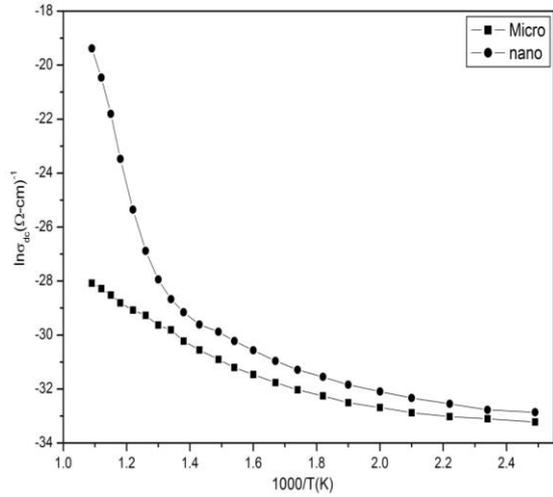
Figure 2: *Temperature dependent DC conductivity.*

From the plot of DC conductivity Vs temperature, it can be concluded that the DC conductivity increases linearly at low temperatures and increases exponentially at high temperatures for both micro and nano samples. At high temperatures the DC conductivity sharply increases for nano sample and is much greater than the increase in the DC conductivity of micro sample. The electrical parameters have been calculated from the least- square straight-line fits using equation (1). We have calculated the DC conductivity and activation energy for both the samples at 550 K and 820 K which are given in the table 1. From the table 1, it is clear that at 550 K, the DC conductivity of the nano sample is nearly two times greater than the DC conductivity of the micro sample and at 820 K it is forty times greater than the DC conductivity of the micro sample. The activation energies of the nano sample at 550 K and 820 K are greater than the activation energies of the micro sample. Although neither the interpretation of σ_0 nor ΔE is straightforward; it is rather ambiguous because electrical conduction involves various transport processes.

Table 1*: Electrical Parameters*

At	T = 550 K			T = 820 K		
	σ_{dc} $(\Omega$ .m)$^{-1}$ x10^{-14}	σ_0 $(\Omega.$ m)$^{-1}$ x10^{-12}	ΔE (eV)	σ_{dc} $(\Omega$ cm)$^{-1}$ $\times 10^{-12}$	σ_0 $(\Omega$ cm)$^{-1}$ $\times 10^{-10}$	ΔE (eV)
Micro	0.99	1.89	0.24	0.24	6.96	0.54
Nano	1.99	64.78	0.36	9.80	64.1	3.21

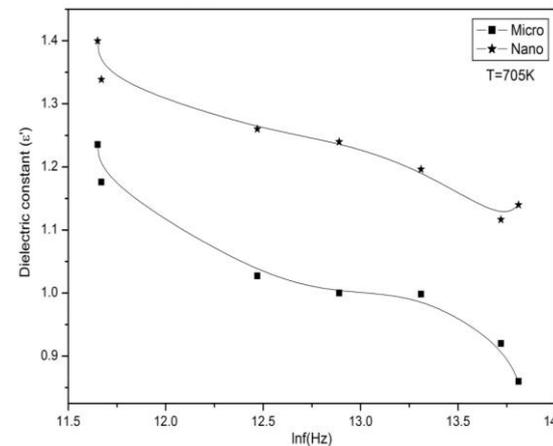## 3.2. Dielectric Constant and Dielectric Loss

In the experimental observations for micro sample, the thickness of pellet (t) has been taken 0.08 cm, radius (r) = 0.5 cm, and C_0 measured 0.4962 pico farad. Similarly for nano sample thickness (t), radius and C_0 measured 0.03 cm, 0.5 cm and 0.9263 pico farad respectively.
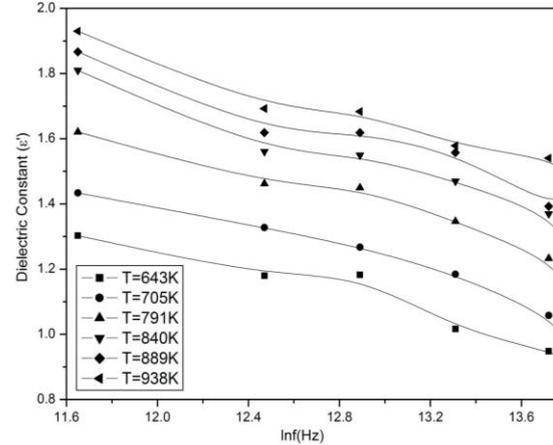
Figure 3 (a): *Frequency dependent Dielectric Constant (ε´) micro sized Si₃N₄ ceramic*
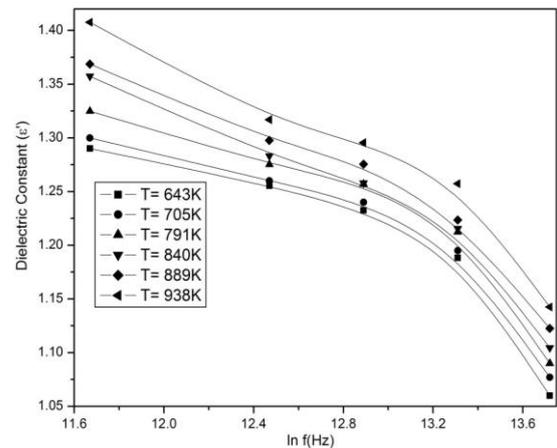
Figure 3 (b): *Frequency dependent Dielectric Constant (ε´) nano sized Si₃N₄ ceramic*

Figure 3 (c): *Frequency dependent Dielectric Constant (ε´) micro-nano sized Si₃N₄ ceramic*

Figure 3 (a,b) shows ε′ vs frequency at different temperatures for micro and nano sample respectively. It is found that the dielectric constant decreases with increase in the frequency ranges from KHz to MHz and sharply decreases for nano sample for the high frequencies. It is also found that the dielectric constant decreases in high frequency range for both micro and nano sample. From the plot of, ε′ vs frequency at constant temperature 705 K for micro-nano sample as shown in figure 3 (c), it is found that the dielectric constant decreases with frequency for both samples but again increases at high frequencies for nano sample. ε″ Vs frequency at different temperatures for micro sample is shown in figure 4 (a) and it is found that there is small continuous decrease in the dielectric loss with increasing the frequency from the KHz range to the MHz range. From the figure 4 (b) of ε″ Vs frequency at different temperatures for nano sample, it is found that the dielectric loss is almost constant for the frequencies in the KHz range and sharply decreases for the frequencies in the MHz range and becomes negative. From the figure 4 (c) of ε″ Vs f at a low temperature (705K) for micro-nano sample it is clear that the dielectric loss is almost constant with frequency for micro sample but it is negative and sharply decreases at high frequencies for nano sample.
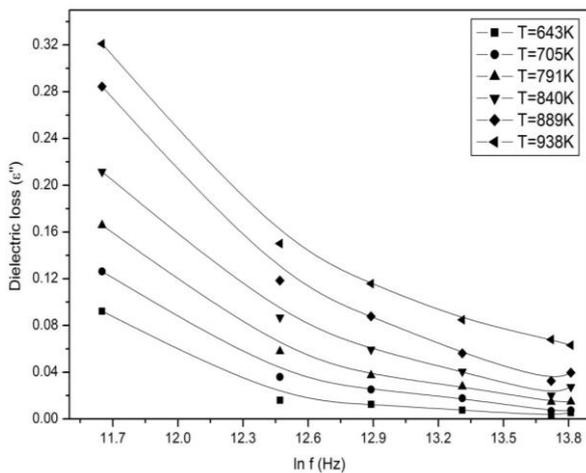

Figure 4 (b): *Frequency dependent Dielectric loss (ε″) nano sized Si$_3$N$_4$ ceramic*
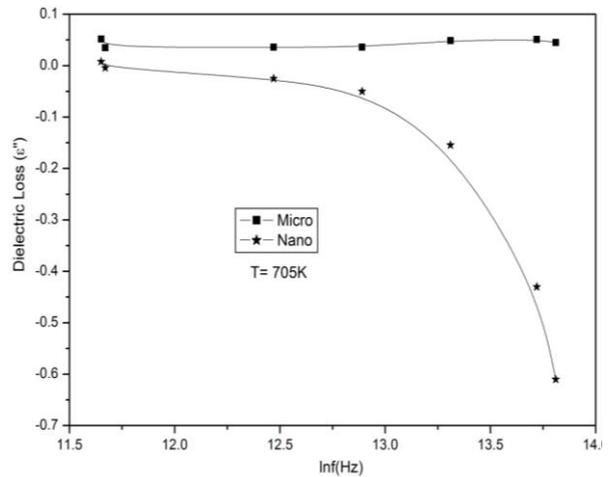
Figure 4 (c): *Frequency dependent Dielectric loss (ε″) micro-nano sized Si$_3$N$_4$ ceramic*

Figure 4 (a): *Frequency dependent Dielectric loss (ε″) micro sized Si$_3$N$_4$ ceramic*
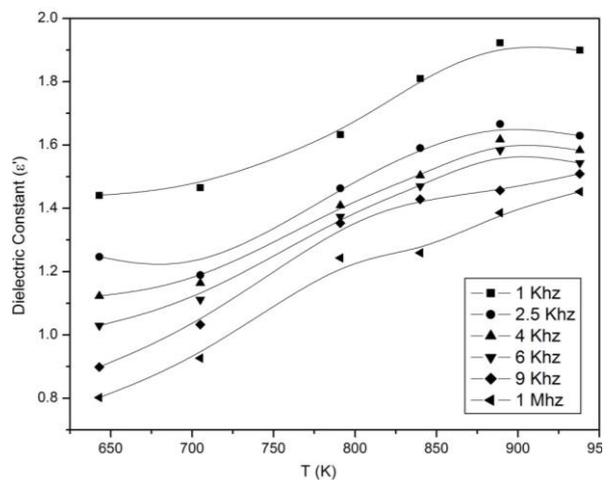
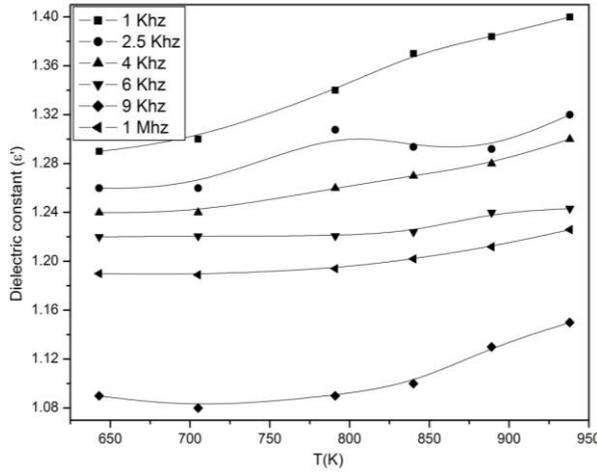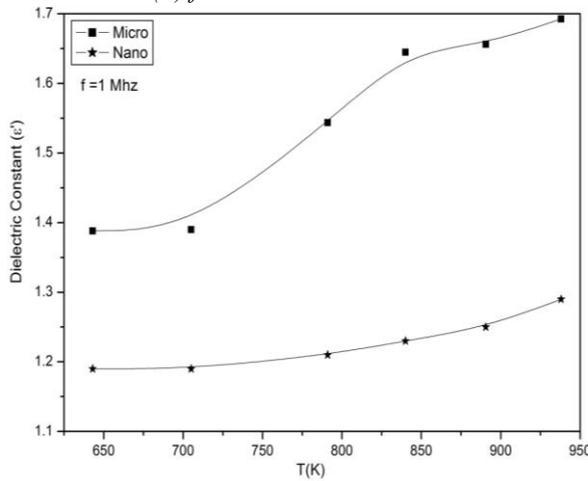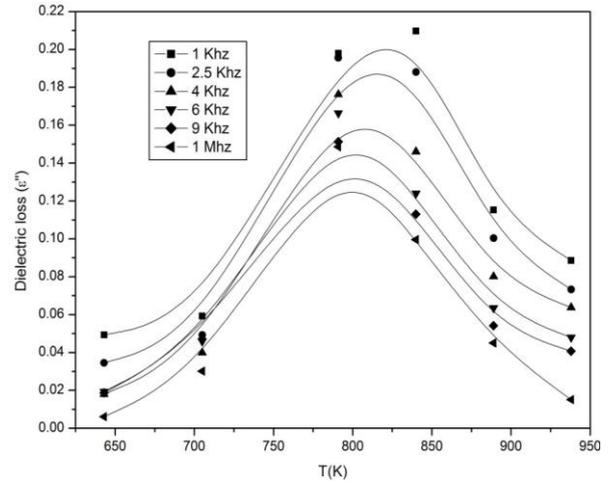**Figure 5 (a):** *Temperature dependent Dielectric constant (ε′) for micro sized Si$_3$N$_4$ ceramic*

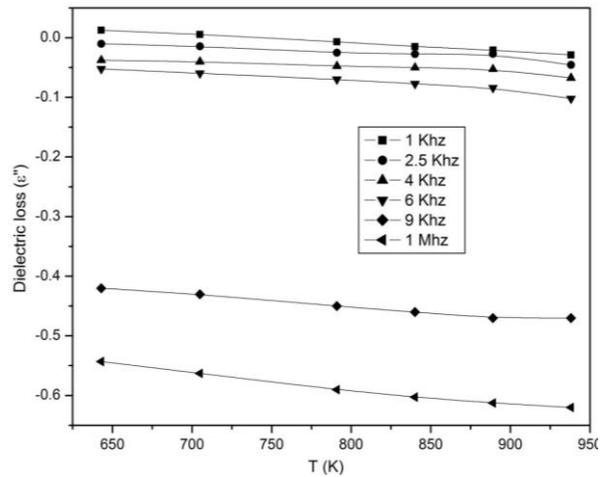**Figure 5 (b):** *Temperature dependent Dielectric constant (ε') for nano sized Si₃N₄ ceramic*

Figure 6 (a): *Temperature dependent Dielectric loss (ε″) for micro sized Si₃N₄ ceramic*

Figure 5 (c): *Temperature dependent Dielectric constant (ε') for nano sized Si₃N₄ ceramic*

Figure 6 (b): *Temperature dependent Dielectric loss (ε″) for nano sized Si₃N₄ ceramic*

From the figure 5 (a) of ε′ Vs T at different frequencies for micro sample, it is found that the dielectric constant is temperature dependent. From the figure 5 (b) of ε′ Vs T at different frequencies for nano sample, it is found that the dielectric constant is almost independent of the temperature but its value is lowered at high frequencies. From the figure 5 (c) of ε′ Vs T at high frequency (1MHz) for micro-nano sample, it is found that the dielectric constant is temperature dependent for micro sample but it is almost constant for nano with temperature and its value is lower than that of micro sample.

From the figure 6 (a) of ε″ Vs T at different frequencies for micro sample it is found that, the dielectric loss is temperature dependent. From the figure 6 (b) of ε″ Vs T at different frequencies for nano sample it is found that, the dielectric loss is almost constant with temperature and its value is lowered for high frequencies. From the figure 6 (c) of ε″ Vs T at a low frequency (1 KHz) for micro-nano sample it is found that the dielectric loss is almost independent of temperature for both micro and nano sample and is negative for nano sample.
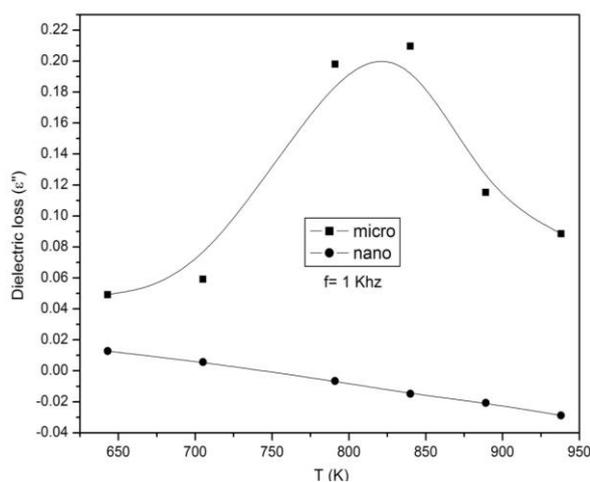
Figure 6 (c): *Temperature dependent Dielectric loss (ε'') for micro-nano sized Si₃N₄ ceramic.*

4. Conclusion

Electrical parameters such as dc conductivity, pre-exponential factor and activation energy are calculated at 550 K and 820 K. Behaviour of dielectric constant and dielectric loss are also observed at different frequencies in the range of 1KHz to 1MHz and at different temperatures in the range of 643K to 938K. Interesting results has been obtained for silicon nitride nano powder. Great impact of grain size on electrical conductivity and dielectric properties of Sintered Silicon Nitride Ceramics has been found. The result shows more efficiency of electrical and dielectric properties with nano grain sizes. The dc conductivity increases exponentially at temperature greater than 800 K. Dielectric constant and dielectric loss are measured in the temperature range 300 K to 900 K with frequency range 1 KHz to 1 MHz. Samples are characterized by the Scanning Electron Microscope (SEM) to confirm its grain size. These types of samples can be used as a high temperature semi-conducting materials.

Acknowledgment

How to Cite this Article:

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