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Influence of Additive Nd_2O_3 on Dielectric Constants for Ternary $Na_2B_4O_7$-SiO_2-Ge_2O_3 System

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ABSTRACT

The dielectric constant and the power factor can be varied by changes in the glass composition. The present work aims to understand the effect of Nd_2O_3 on dielectric properties of $Na_2B_4O_7$-SiO_2-Ge_2O_3 system. The results show that, ln σ, exponent factor S, ε′ and ε″ values of $Na_2B_4O_7$-SiO_2-Ge_2O_3 increased with increasing Nd_2O_3 content. ε′and ε″ values of $Na_2B_4O_7$-SiO_2-Ge_2O_3 increased with increasing temperature. Activation energy, the exponent factor S and M′ values of $Na_2B_4O_7$-SiO_2-Ge_2O_3 deceased with increasing Nd_2O_3 content. The exponent factor S for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ decreased with increasing temperature. M′ value decreased with increasing temperature, but it increased with increasing frequency.

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Introduction

Amorphous solids are compounds without long range order and periodicity, where glasses are a subset of its which show a distinct glass transition temperature made by solidifying molten silicon dioxide with other chemical compounds. Glasses established significant attention due to their sole properties such as hardness, excellent corrosion resistance, good strength and transparency. The structure of glass through many years studied by infrared spectroscopy, differential scanning calorimetry and x-ray diffraction analysis [1-4]. Transition metal oxides have various interesting properties as superconductivity, magneto resistance, fiber optic communication devices, piezoelectricity solid-state lasers luminescent and solar energy concentrators [5]. Some research for the SiO_2 -B_2O_3 -Na_2O ternary system have shown breaking down the original network, due to that alkali cations (Na^+) can played as charge compensators in the four coordinated boron [6, 7]. The aim of this work is to study the effect of Nd_2O_3 on dielectric properties of $Na_2B_4O_7$-SiO_2-Ge_2O_3 system.

Experimental Details

$Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ (x=0, 1, 1.5 and 2.5 mol. %) samples, Table 1, were prepared by the conventional melt quench technique, with high purity (99.9%) components powders, obtained from Sigma Aldrich. A pure regent of the starting materials is mixed with the proper ratios. The mixture for each sample was well grinded to insure the homogeneity of the powder. The mixture powder was melted in porcelain crucible at 1200 °C for one hour. The molten mixture was stirred gradually to ensure the homogeneity. The molten samples were poured and quenched between two brass plates. The produced samples were annealed for 2 hours to reduce thermal stresses inside the samples. The dielectric measurements of used glasses were carried out by means of a Novocontrol high resolution alpha dielectric analyzer in the frequency range 0.01 Hz to 10 MHz. The analyzer was supported by Quatro temperature controller using pure nitrogen as heating agent and providing a temperature stability better than 0.2 K. The measurements were conducted using gold-plated stainless-steel electrodes of 20 mm in diameter in parallel plate capacitor configuration.

Table 1: Composition of Used Samples

Samples	$Na_2B_4O_7$	SiO_2	Ge_2O_3	Nd_2O_3
G1	70	15	15	0
G6	70	15	14	1
G7	70	15	13.5	1.5
G8	70	15	13	2
G9	70	15	12.5	2.5

Results And Discussions

Dielectric Constant

Figure 1 shows the relation between ln σ and activation energy versus Nd_2O_3 concentration for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$. It is obvious that, ln σ value increased but of the dc conductivity activation energy deceased with increasing Nd_2O_3 content. Also Figure 2 shows the relation between ln σ and 1000/T for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ at different frequencies. The results show ln σ increased with increasing frequency and temperature. hat is may be attributed to the partial conversion of BO_4 to BO_3 structural units with the formation of non-bridging oxygen atoms, because the electrons are less tightly bound to the nuclear charge and easily excite from the valence to conduction band. Moreover, the presence of Nd ions affects the network structure increasing the number of free electrons leading to increase conductivity.

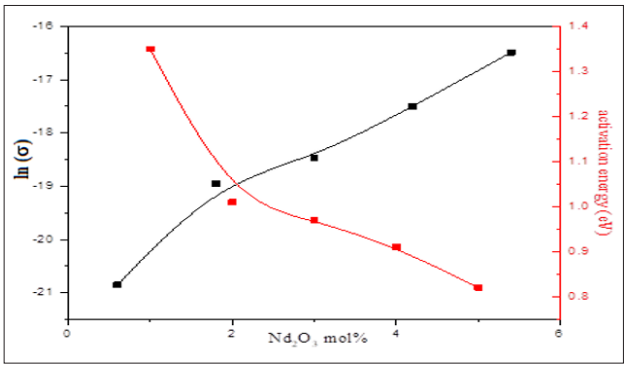

Figure 1: ln σ and Activation Energy Versus Nd_2O_3 Concentration for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$

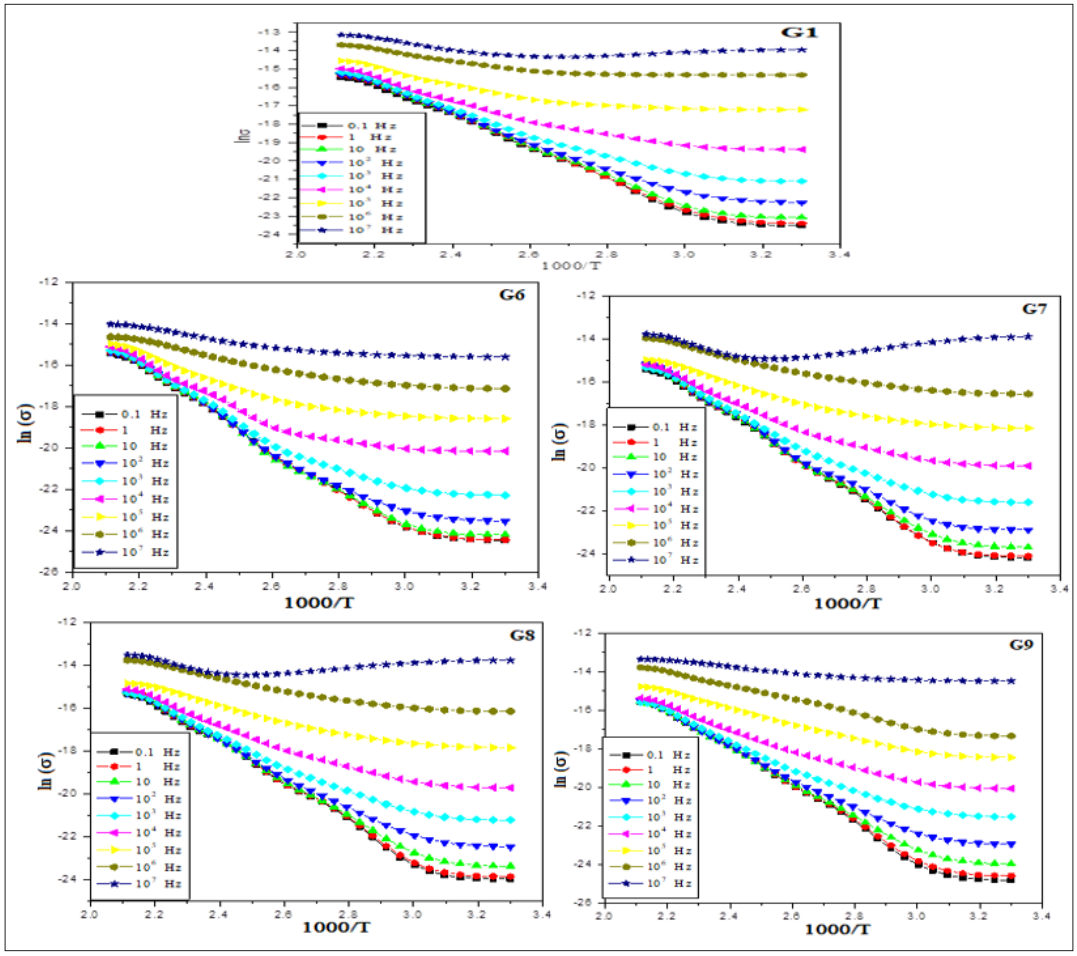

Figure 2: ln σ versus 1000/T for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ at Different Frequency

The frequency dependence of the total conductivity σ(ω) of all amorphous materials and glasses follows relation:

$$\sigma(\omega) = \sigma_{dc}(0) + A\,\omega^s$$

Where σ_{dc} (0) is the frequency independent conductivity, A is a constant, S is the exponent factor and ω is the angular frequency. The dielectric loss factor describes the loss of electric field energy in the material. Therefore, the exponent s was obtained by plot log σ_{total} versus log ω. The relation between the exponent factor S and T for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$, Figure 3, shows it decreased with increasing temperature and increased with increasing Nd_2O_3 content.

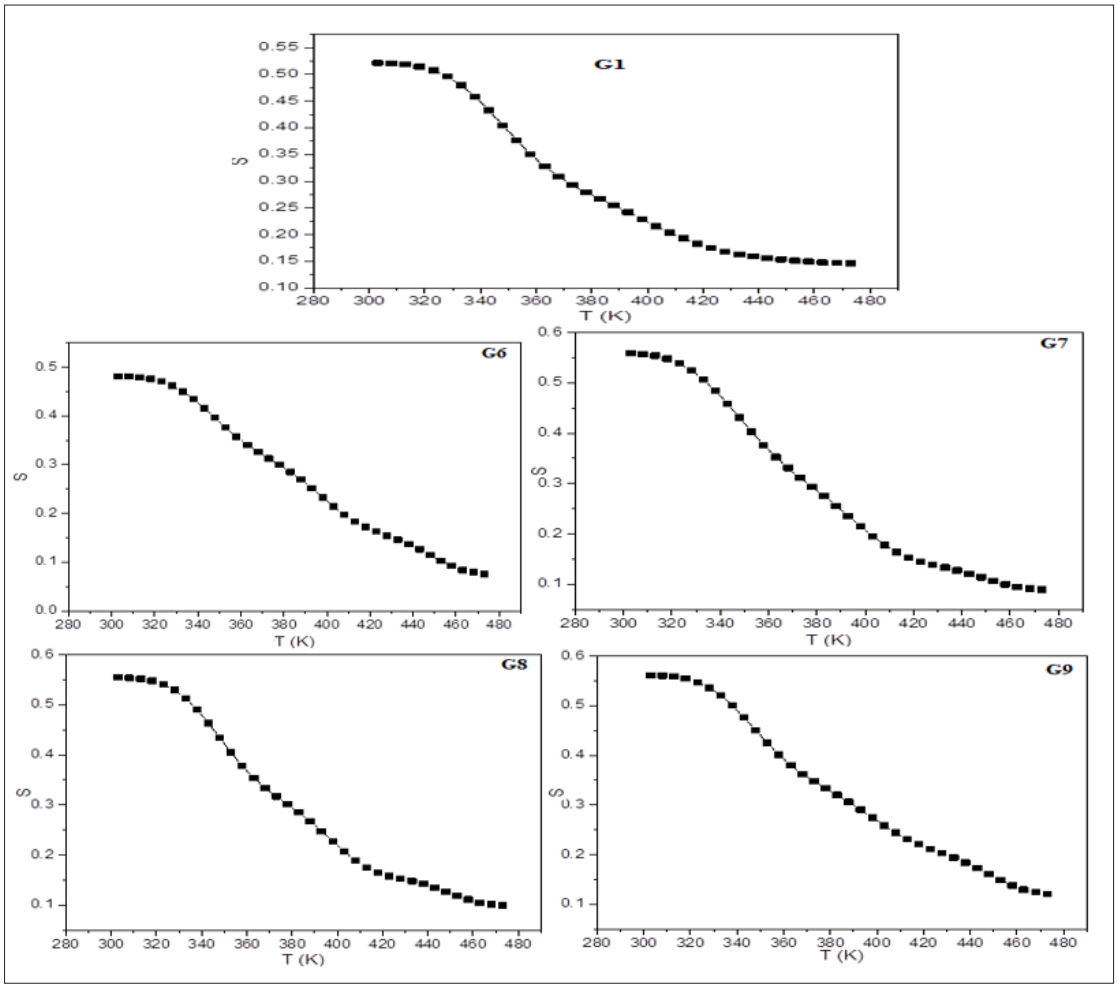

Figure 3: Exponent Factor S, versus T for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$

The dielectric constant was measured in the frequency range 0.01 Hz up to 10 MHz. The value of dielectric loss (ε′) for $Na_2B_4O_7$-SiO_2-Ge_2O_3 increased after Nd_2O_3 adding content as shown in Figure 4. The relation between dielectric constant (ε′) and temperature for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$, Figure 5, shows ε′ increased with increasing at low frequency with a slight or no change at high frequency. The value of the dielectric loss factor (ε″) increased with increasing Nd_2O_3 concentration in $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ as shown in Figure 6. The relation between ε″ and temperature for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ shown in Figure 7, shows ε″ increased with increasing temperature at low frequency but increased slowly with increasing temperature at high frequency. That is meant, the variation of the dielectric constant is large at low frequency and vice versa, which can be attributed to the orientational polarization is related to the thermal motion of molecules [8] and/or the space charge polarization due to the bonding defects in the structure [9]. The dielectric constant decrease with increasing frequency is a normal dielectric behavior where that agreed by other researchers [10-12].

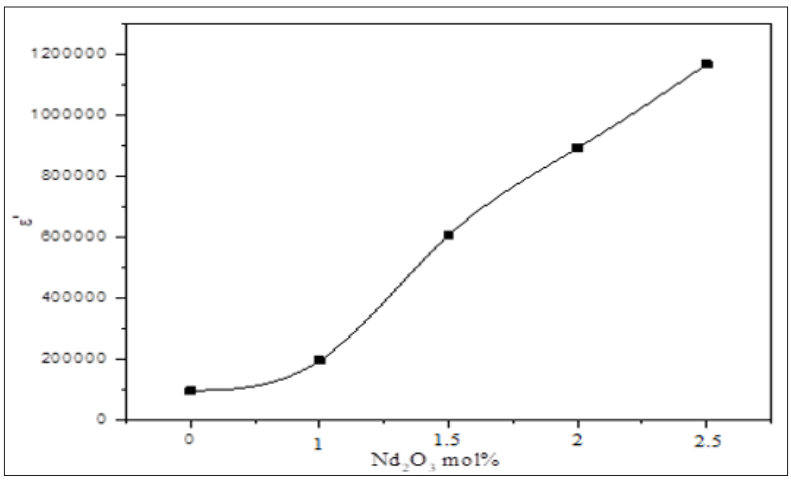

Figure 4: ε′ Versus Nd_2O_3 Concentration

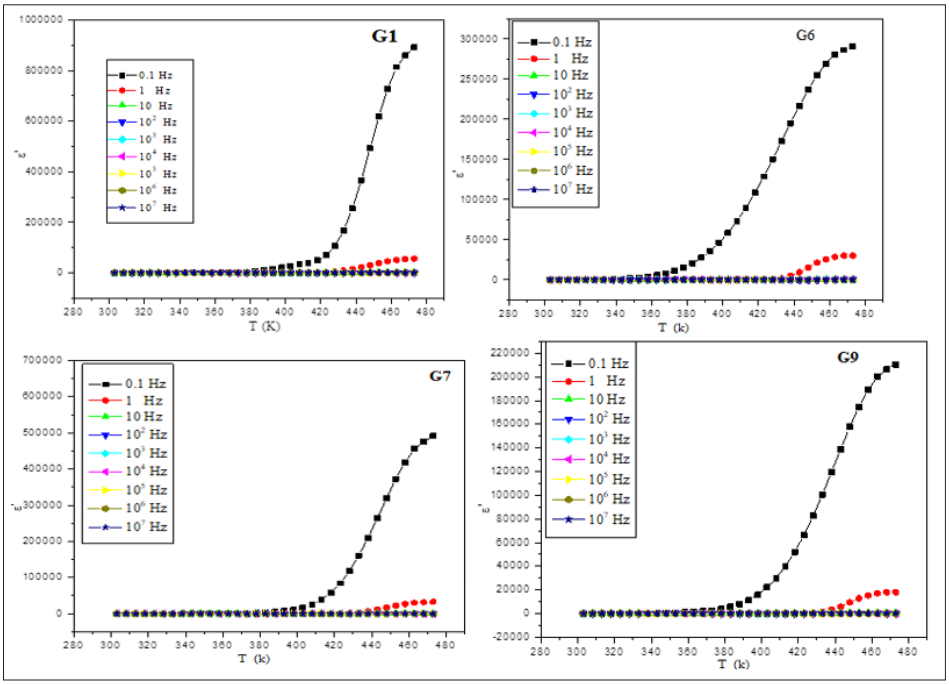

Figure 5: ε′ Versus Temperature for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ at Different Frequency

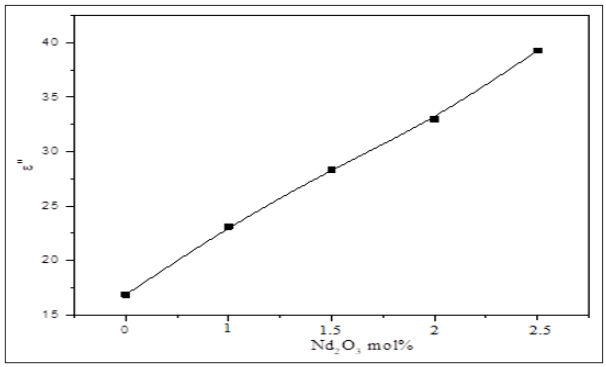

Figure 6: ε″ versus Nd_2O_3 Concentration

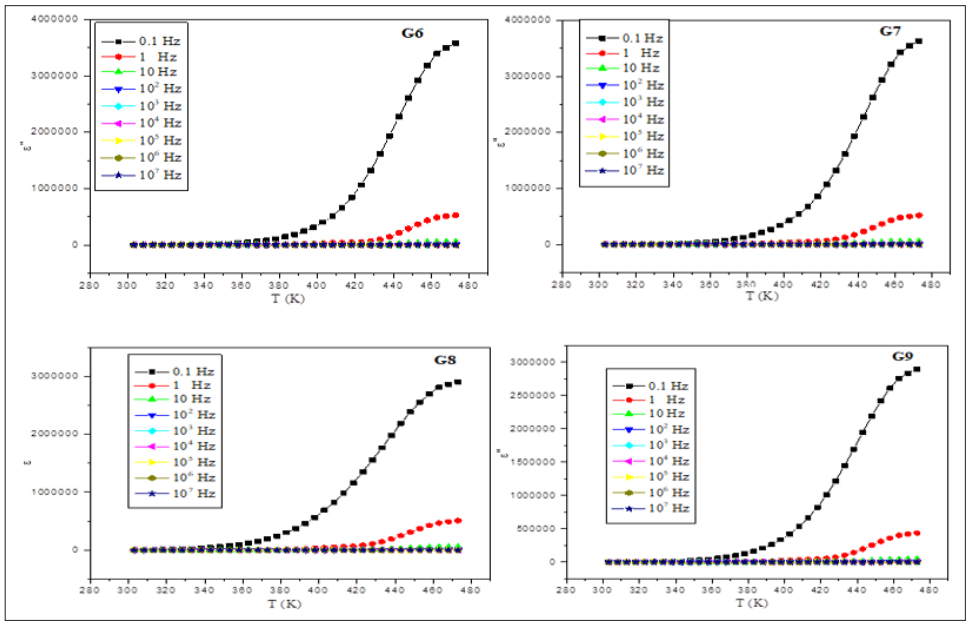

Figure 7: ε″ Versus Temperature for $Na_2B_4O_7$-SiO_2-$(Ge_2O3)_{2.5-x}$-$(Nd_2O_3)_x$

The M′ value decreased with increasing Nd_2O_3 concentration in $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ as shown in shown Figure 8. Also, the relation of M′ versus temperature for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$ shows, it decreased with increasing temperature, but it increased with increasing frequency.

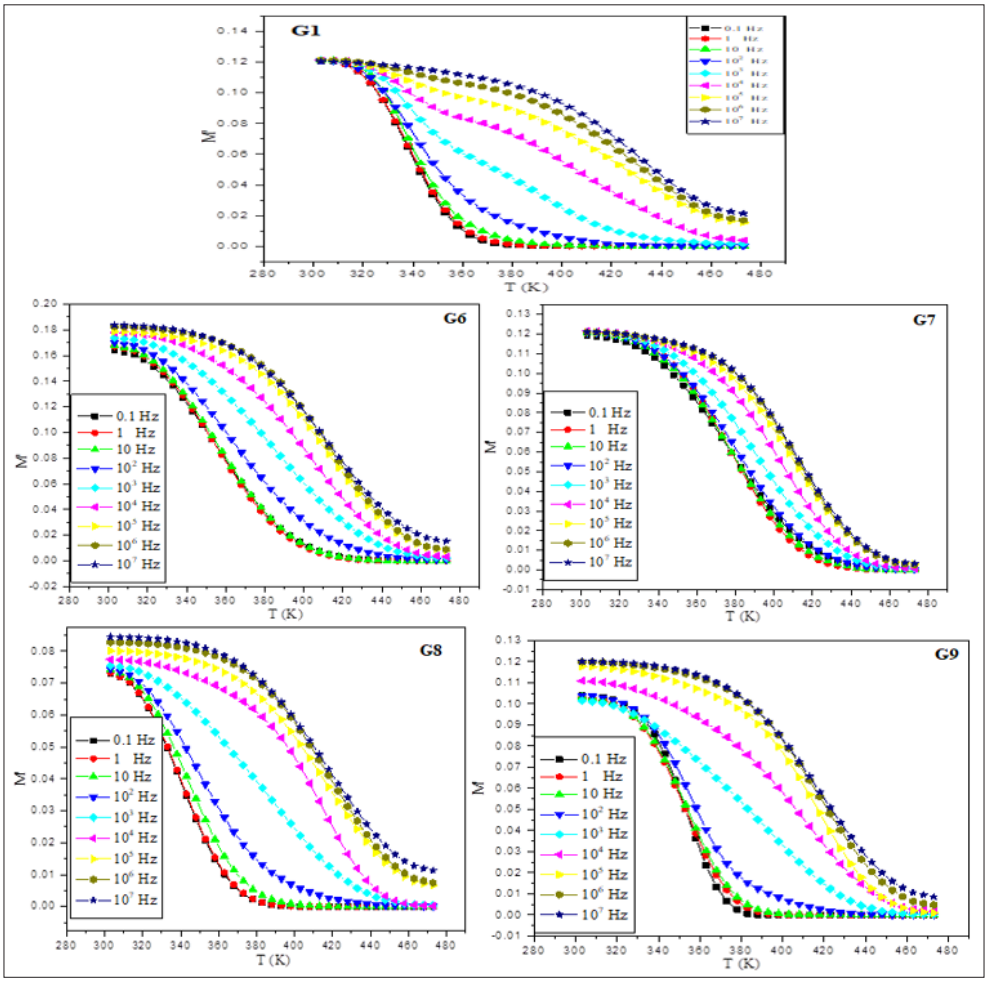

Figure 8: M′ Versus Temperature for $Na_2B_4O_7$-SiO_2-$(Ge_2O_3)_{2.5-x}$-$(Nd_2O_3)_x$

Conclusion

Our 11B NMR spectra results show that, adding Nd_2O_3 to $Na_2B_4O_7$-SiO_2-Ge_2O_3 decreased the degree of connectivity and breaking of Si-O-Si bond, and formed anti-symmetric stretching of Si-O-Nd, therefore, Nd_2O_3 enter network as a modifier and affected the probability of N_4 formation causing a change in dielectric constants.

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