

**Research Article**

## **DIELECTRIC-SPECTROSCOPIC AND A.C. CONDUCTIVITY INVESTIGATIONS ON COPPER DOPED $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$ CERAMICS**

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### **ABSTRACT**

The dependence of loss tangent ( $\tan\delta$ ) on temperature and frequency has been reported for Sodium Lithium trititanate ( $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$ ) ceramic and its 0.01, 0.1 and 1.0 molar percentage of CuO doped derivatives. The losses are the characteristic of dipole mechanism and of electrical conduction. The dependence of dielectric constant ( $\epsilon'$ ) on temperature and frequency has further been reported for these samples. The dependence of dielectric constant ( $\epsilon'$ ) on frequency and temperature indicates the presence of less aligned electric dipoles in the interlayer space in the lower temperature range. The rise in temperature furnishes the orientation as well as dissociation of dipoles. For further rise in temperature, the degree of orderliness of their orientation is diminished. In dielectric constant versus temperature plots the existence of broad peak in higher temperature region may be due to the possible ferroelectric's phase transition. The results of a.c. conductivity in temperature range 373K-773K have also been reported in this chapter. The  $\text{Ln}(\sigma T)$  versus  $1000/T$  plots have been divided into four regions, Region-I, Anomalous region, Region-II and Region-III. The various conduction mechanisms in the different regions have been stressed in this chapter. Furthermore, the  $\text{Ln}\sigma T$  versus frequency plots for all the above samples reveal that the electronic hopping conduction (polaron) is dominant in the lower temperature region.

### **INTRODUCTION**

Advanced ceramic materials constitute a technology with a very broad base of current and potential applications and a growing list of material compositions.[Advanced ceramics and nano ceramics 2003] Some new potential dielectric materials have been made through the cross-substitution of  $\text{Ba}^{2+}$  by 1:1 molar combinations of tri ( $\text{La}^{3+}$ ) and monovalent ( $\text{Li}^+$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ) at the  $\text{Ba}^{2+}$  site in  $\text{BaTiO}_3$ . [Uchida 1997] The impedance spectra of Nb doped  $\text{SrTiO}_3$  ceramic suggests that under oxidizing condition, the overall electrical behavior is determined by two microstructural contribution.[Abrantes 2002] Synthesis and dielectric characteristic of layered  $\text{Bi}_{4-x}\text{R}_x\text{Ti}_3\text{O}_{12}$  has been studied by Jose L. Pineda.[Jose 2003] A series of layered perovskite with general formula  $\text{A}_2\text{Ln}_2\text{Ti}_3\text{O}_{10}$  (where A= Na, K, Rb and Ln= La, Nd, Gd, Dy) has been synthesized by many researchers.[Gondrand 1987, Gopalakrishn 1987] Very recently alkali titanates  $\text{A}_2\text{Ti}_n\text{O}_{2n+1}$  (A= Li, Na, K) were synthesized by hydrothermal reaction of titania powder in an aqueous alkaline solution.[Nagase 1999, Yamawaki 2000, Masaki 2000, Tomiha 2002] The a. c. conductivity studies in polycrystalline samples  $\text{Na}_2\text{Ti}_3\text{O}_7$  and  $\text{K}_2\text{Ti}_4\text{O}_9$  and their Nb containing products were reported by Kikkawa et al.[Kikkawa 1985] in the temperature range 500K to 730K. The ion exchange reactions involving polycrystalline samples of  $\text{Na}_2\text{Ti}_3\text{O}_7$  and  $\text{K}_2\text{Ti}_4\text{O}_9$  are quite well known [Izawa 1982, Fujiki 1977] The crystal structure of  $\text{Na}_2\text{Ti}_3\text{O}_7$  and  $\text{K}_2\text{Ti}_4\text{O}_9$  do suggest possibility of ionic conduction along the layers and thus application of these materials as ion-exchanger.[Fujiki 1977] Very recently, formation mechanism of  $\text{H}_2\text{Ti}_3\text{O}_7$  nanotubes have been reported by S.Zhang et al.[Zhang 2003] Recently M. Machida et al. reported the studies on pillaring and photocatalytic property of partially substituted layered titanates  $\text{Na}_2\text{Ti}_{3-x}\text{M}_x\text{O}_7$  and  $\text{K}_2\text{Ti}_{4-x}\text{M}_x\text{O}_9$  (M= Mn, Fe, Co, Ni, Cu)[Machida 2000] The crystal structure of  $\text{Na}_2\text{Ti}_3\text{O}_7$  do suggest possibility of ionic conduction along the layers & thus the application of this material as ion exchanger. [Watkins 1959]. Recently the refinement of crystal structure of  $\text{Na}_2\text{Ti}_3\text{O}_7$  has been done by V.V. Kireev [Kireev 2003]. Lithium diffusion in nano crystalline

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composite system  $(1-X) \text{Li}_2\text{O} : x\text{Al}_2\text{O}_3$  was studied by NMR measurements [Wilkening 2003]. In assessing the possibility of high ionic conduction needed for battery application various Li-containing materials, particularly those with structure arrangements containing tunnels or diffusion paths have been examined. The conductivity generally decreases with increasing lattice concentration. It is agreeable with lithium ion conductivities in  $\text{Ln}_{0.5}\text{Li}_{0.5}\text{TiO}$  [Mohanram 1987] are lowered with decreasing the radii of lanthanide ions (from  $\text{Ln}=\text{La}$  to Pr, Nd & Sn) [Itoh 1994]. On the other hand the increase of sodium content in  $[(\text{La}_{0.5}\text{Li}_{0.5})(1-X)(\text{La}_{0.5}\text{Na}_{0.5}X)\text{TiO}]$  [Mohanram 1987] lowers the ionic conductivity in spite of the lattice expansion. Physico-chemical properties of alkali titanates such as optical properties, ionic and electronic conductivities [Machida 2000, Shripal 1991] have been investigated on account of their application in the industry as ion exchangers, electrodes for secondary batteries, filters, reinforcement, heat insulators. Furthermore, for the first time frequency and temperature dependence of the loss tangent ( $\tan\delta$ ) to characterize  $\text{Na}_2\text{Ti}_3\text{O}_7$  and  $\text{NaKTi}_3\text{O}_7$  were reported by Shripal et al. [Shripal 1990].  $\text{Li}_2\text{Ti}_3\text{O}_7$  with the ramsdelite ( $\text{M}_n\text{O}_2$ ) structure was reported as a superionic conductor having an isotropic conductivity along the channel in the structure [Boyce 1979]. Layered poly type of  $\text{Li}_2\text{Ti}_3\text{O}_7$  may be prepared by ion exchange of  $\text{Na}_2\text{Ti}_3\text{O}_7$  [Jose 2003]. Shripal et al. have reported the dielectric-spectroscopic and a. c. conductivity studies of pure and manganese doped  $\text{Na}_2\text{Ti}_3\text{O}_7$ . [Pal Dharmendra 2010, Pal Dharmendra 2010] EPR and electrical studies in layered  $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$  and its Copper and manganese doped derivatives have been reported by Dharmendra Pal et al. [Pal D. 2005, 2004] Accordingly, it seems to be very interesting to characterize  $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$  with 0.01, 0.1 and 1.0 molar percentage of CuO doped derivatives denoted as SLT, CSLT-1, CSLT-2 and CSLT-3 through dielectric-spectroscopic and a. c. conductivity investigations.

## MATERIALS AND METHODS

The ceramic sample  $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$  (denoted as SLT) and its copper-doped derivatives with different molar percentage of CuO (0.01, 0.1 and 1.0) (denoted as CSLT-1, CSLT-2 and CSLT-3) have been synthesized using sintering process. The method of preparation of palletized ceramic samples is similar to that reported earlier in the literature.[23] XRD-patterns for all the compositions were collected by Iso-Debyeflex 2002, Richseifert and Co. diffractometer using  $\text{CuK}\alpha$  radiation generated at 30 kV and 20 mA. The formation of these titanates (SLT, CSLT-1, CSLT-2 and CSLT-3) is confirmed by the XRD-patterns obtained at room temperature (RT).

The flat faces of the sintered pellets were painted with an air-dried high purity silver paste and then mounted in the sample holder evacuated up to  $10^{-3}$  mbar for the electrical measurements. The loss tangent ( $\tan\delta$ ) and parallel capacitance ( $C_p$ ) of the palletized samples were directly measured as a function of temperature and frequency by the HP 4194A impedance analyzer. The relative permittivity ( $\epsilon_r$ ) and the bulk a. c. conductivity ( $\sigma$ ) of the samples were calculated by using expressions-

Dielectric Constant (real)	$\epsilon_r = C_p/C_o$	(i)
Conductance	$G = \omega C_p \tan\delta$ ; where $\omega = 2\pi f$	(ii)
Conductivity	$\sigma = G(t/A)$	(iii)

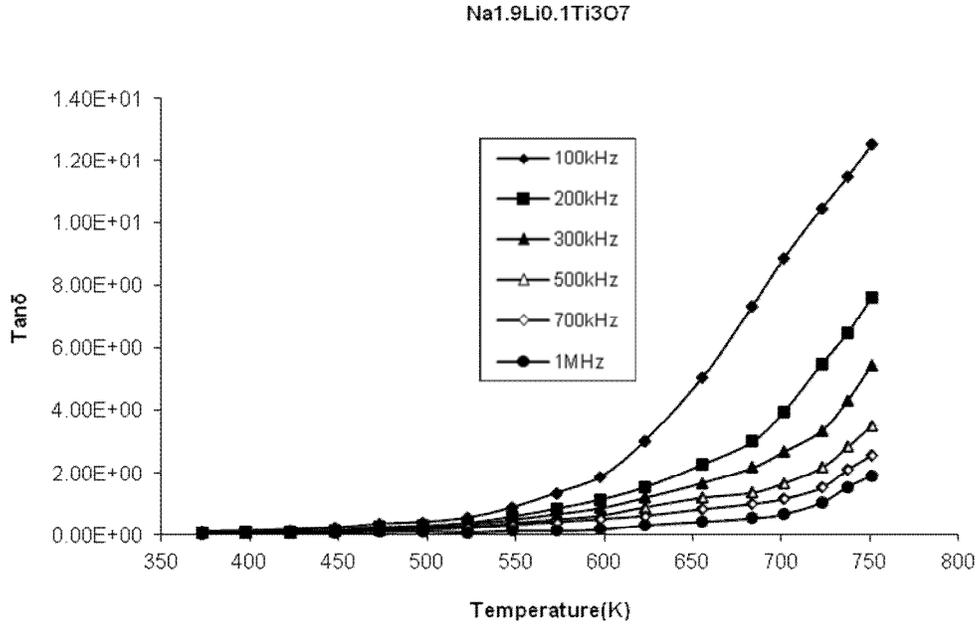
Where  $f$  is the frequency of a. c. signal,  $t$  is the thickness and  $A$  is the area of cross-section of the pellet.

## RESULTS AND DISCUSSION

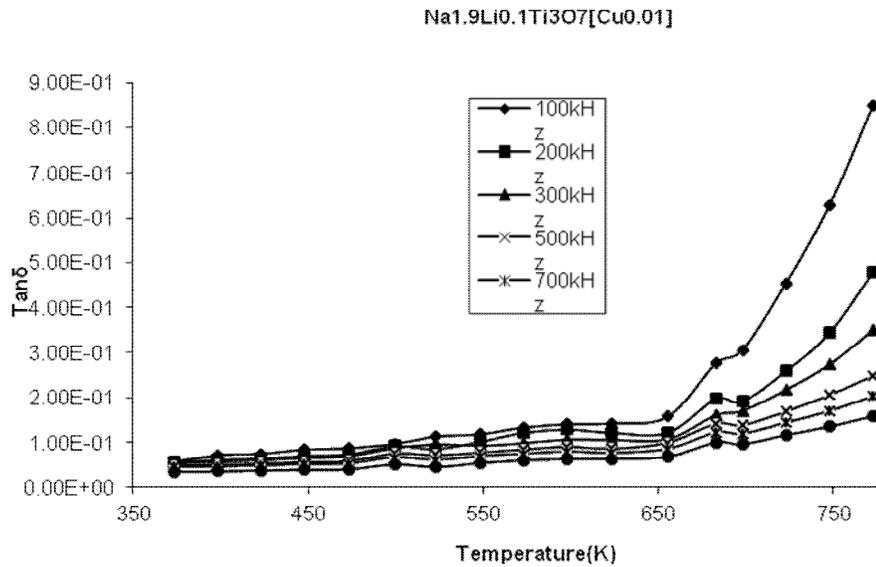
Fig.1 (a), (b), (c), (d) shows the  $\tan\delta$  versus temperature curves for pure and its 0.01, 0.1 and 1.0 molar percentage of CuO doped derivatives in temperature range 373-773K at some fixed frequencies. It can be seen from these plots that the values of  $\tan\delta$  remains invariant with the rise in temperature up to 523K for SLT, 648K for CSLT-1 and CSLT-2 and 548K for CSLT-3 with a small dip at 523K for all the doped derivatives. At higher temperature it can be seen that the rate of increase of  $\tan\delta$  is higher for lower frequencies. This may be due to space charge polarization.[Das 2003] Furthermore, the a frequency independent relaxation peak at 673K is found in  $\tan\delta$  versus temperature curves for all the doped derivatives. The existence of this relaxation peaks can be explained by proposing that the presence of

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dipoles generated in the interlayer space because of one type of copper substitution.  $\text{Cu}^{2+}$  at  $\text{Ti}^{4+}$  sites seem to responsible for creation of the dipoles in the interlayer space giving peak at 673K. The general increase of  $\tan\delta$  with temperature may be due to no. of ions that take part in relaxation polarization continuously grows with the rise in temperature [Kumar 2005].

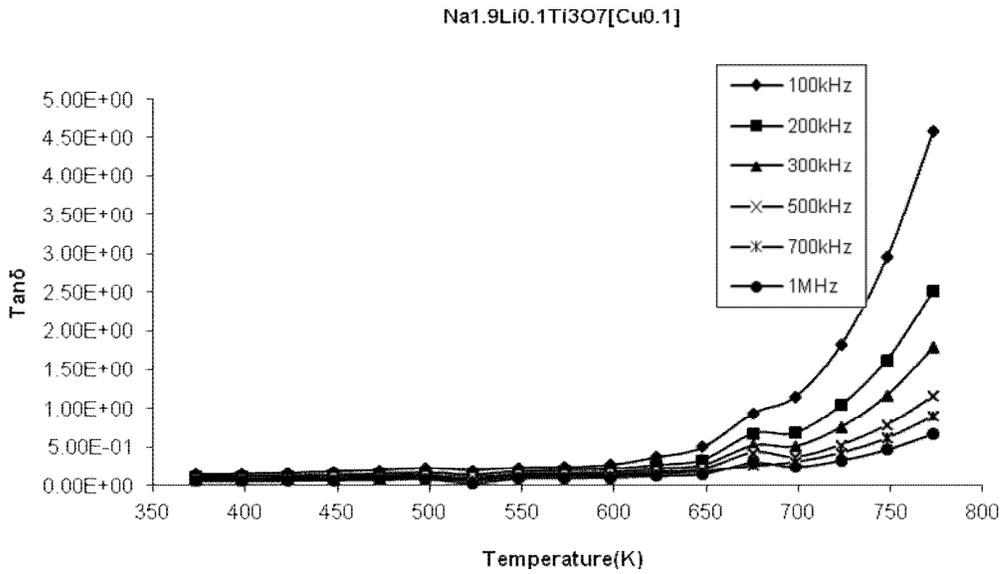


**Figure 1(a). Loss tangent ( $\tan\delta$ ) versus Temperature for SLT**

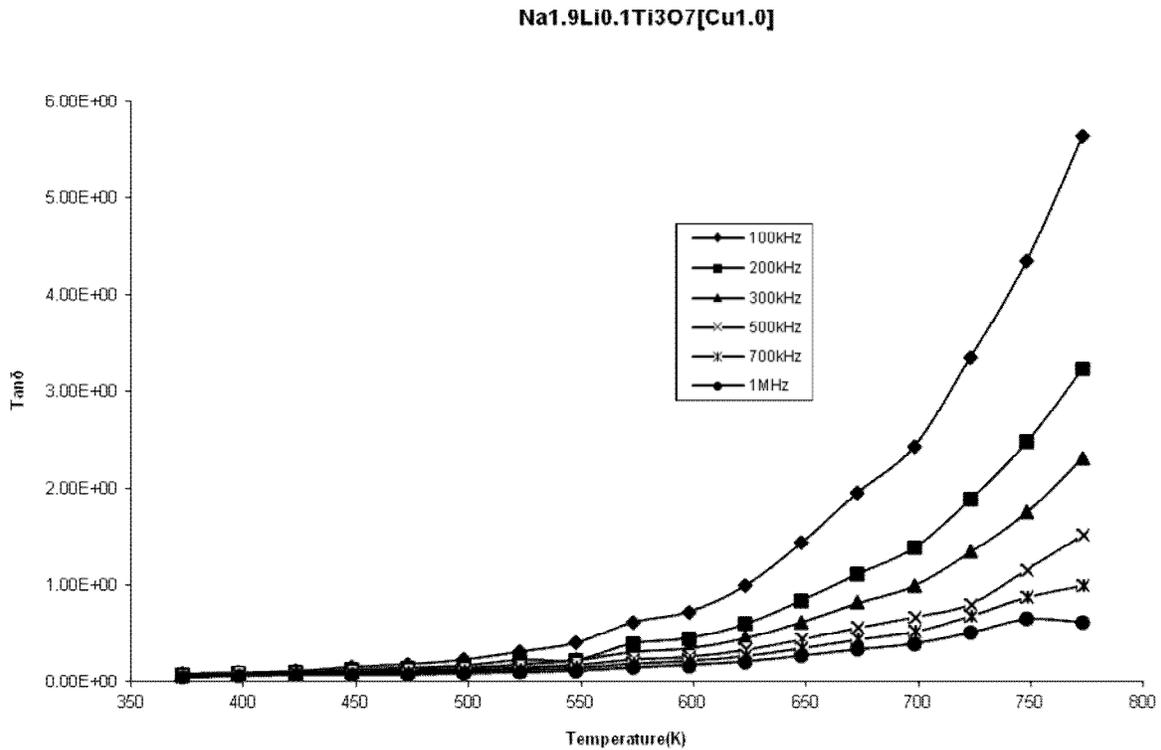


**Figure 1(b). Loss tangent ( $\tan\delta$ ) versus Temperature for CSLT-1**

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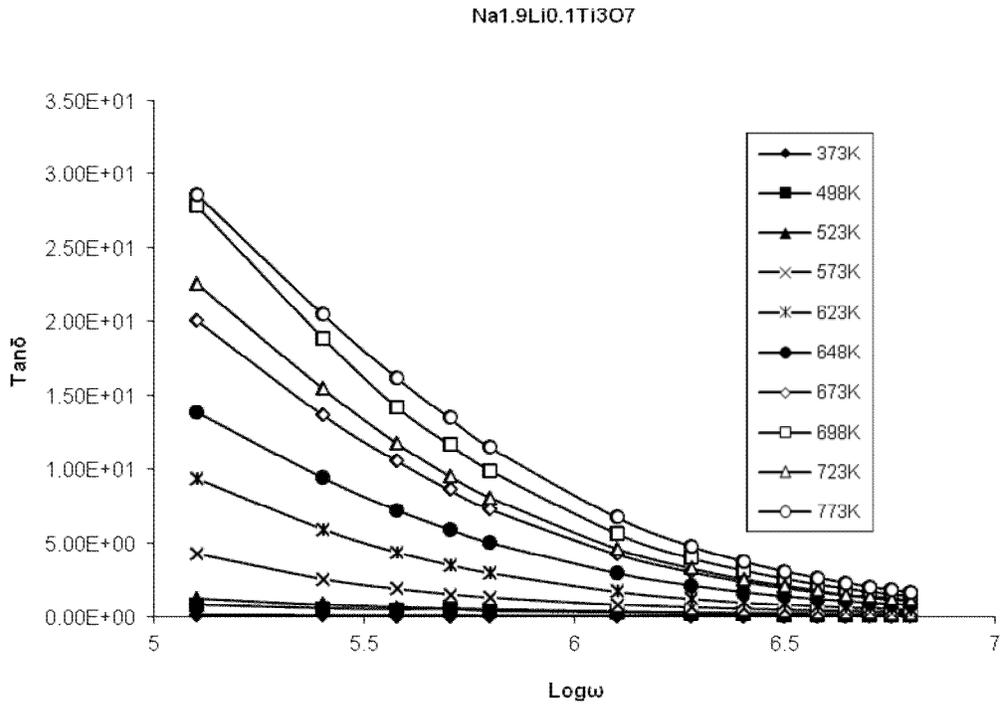


**Figure 1(c). Loss tangent ( $\tan\delta$ ) versus Temperature for CSLT-2**

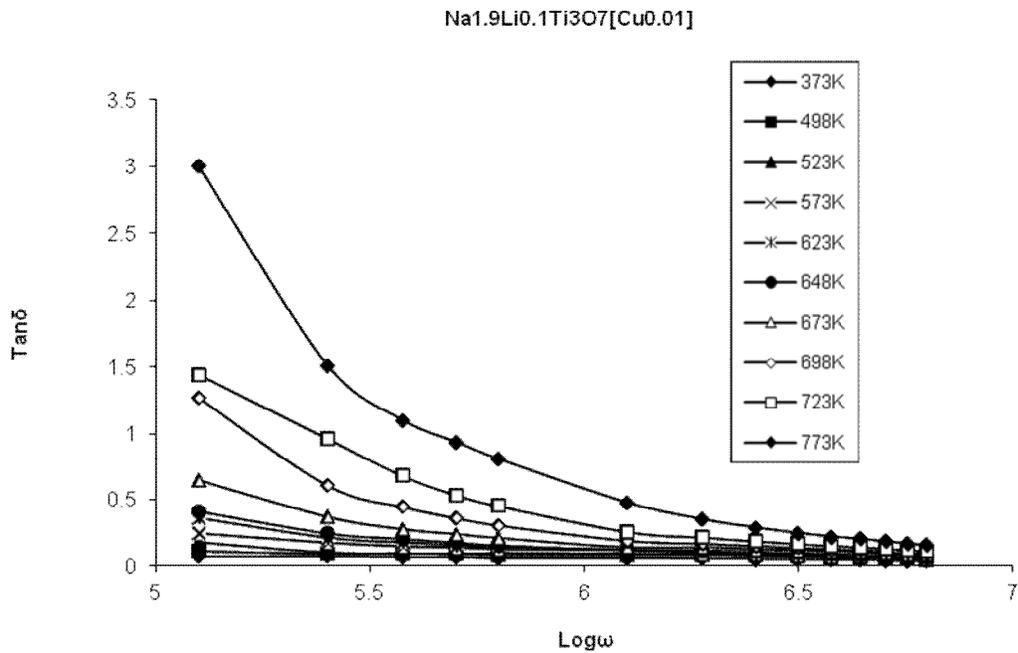


**Figure 1(d). Loss tangent ( $\tan\delta$ ) versus Temperature for CSLT-3**

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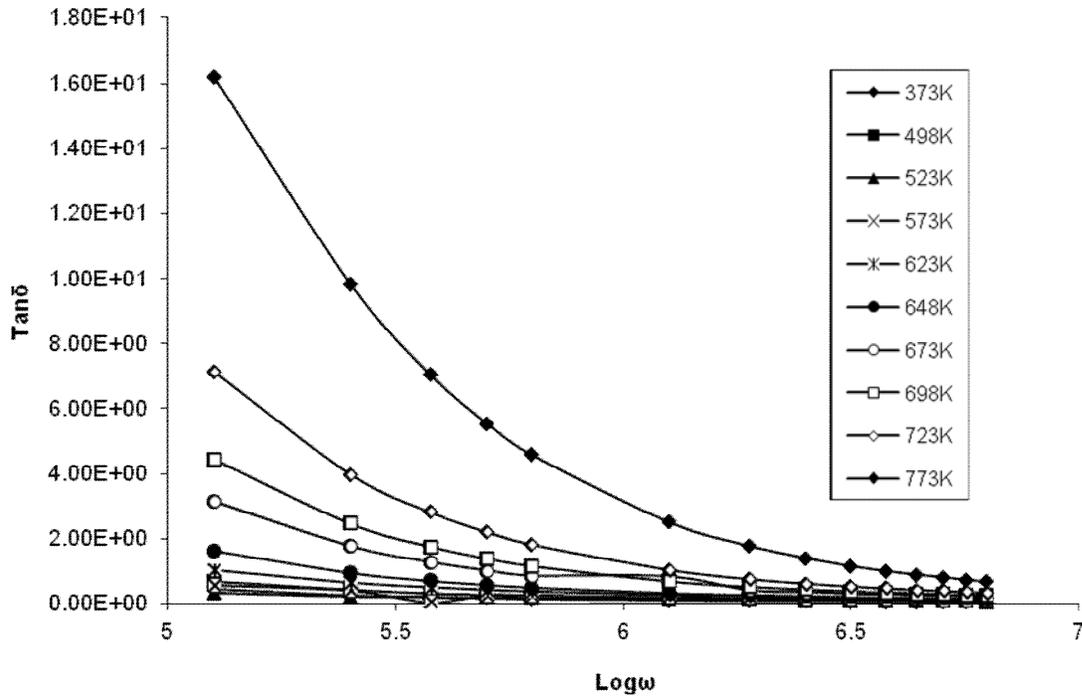
**Figure 2(a). Loss tangent ( $\tan\delta$ ) versus frequency for SLT**



**Figure 2(b). Loss tangent ( $\tan\delta$ ) versus frequency for CSLT-1**

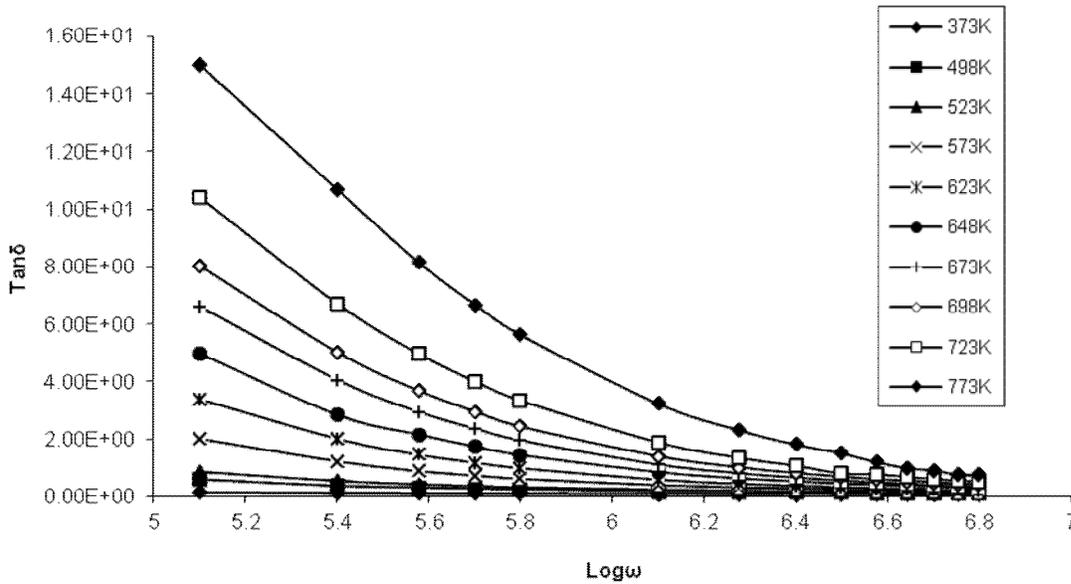
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**Na<sub>1.9</sub>Li<sub>0.1</sub>Ti<sub>3</sub>O<sub>7</sub>[Cu<sub>0.1</sub>]**



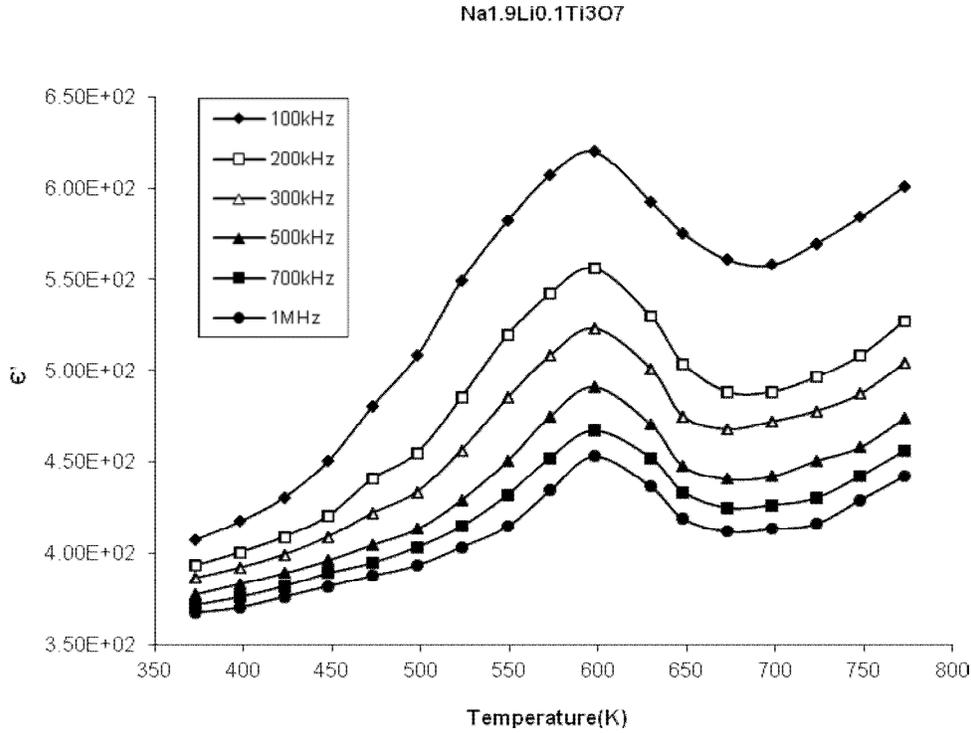
**Figure 2(c). Loss tangent (tanδ) versus frequency for CSLT-2**

**Na<sub>1.9</sub>Li<sub>0.1</sub>Ti<sub>3</sub>O<sub>7</sub>[Cu<sub>1.0</sub>]**

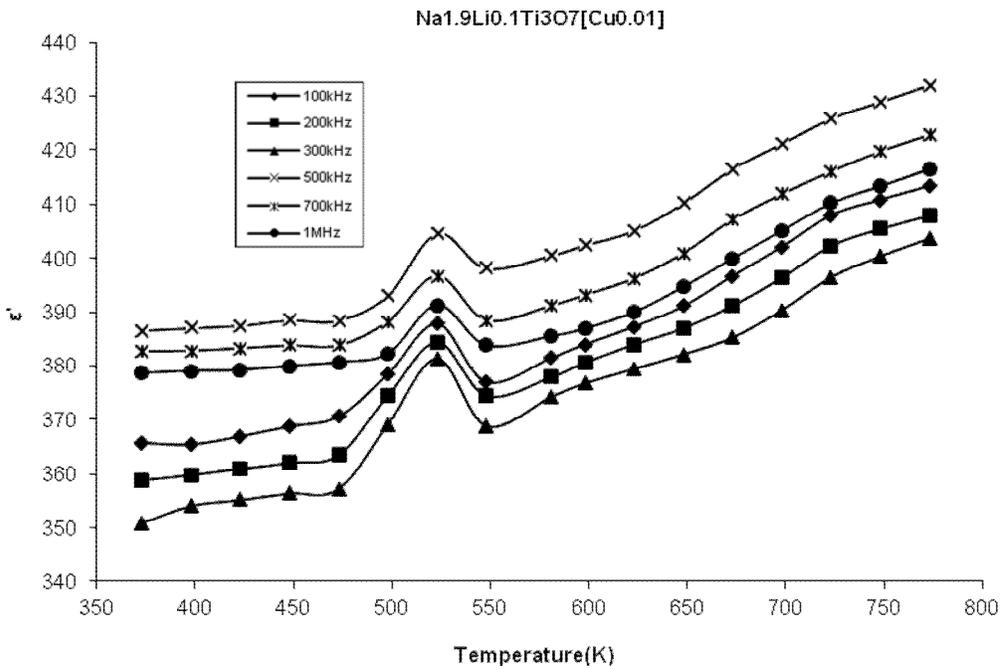


**Figure 2(d). Loss tangent (tanδ) versus frequency for CSLT-3**

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**Figure 3(a). Dielectric constant ( $\epsilon'$ ) versus temperature for SLT**



**Figure 3(b). Dielectric constant ( $\epsilon'$ ) versus temperature for CSLT-1**

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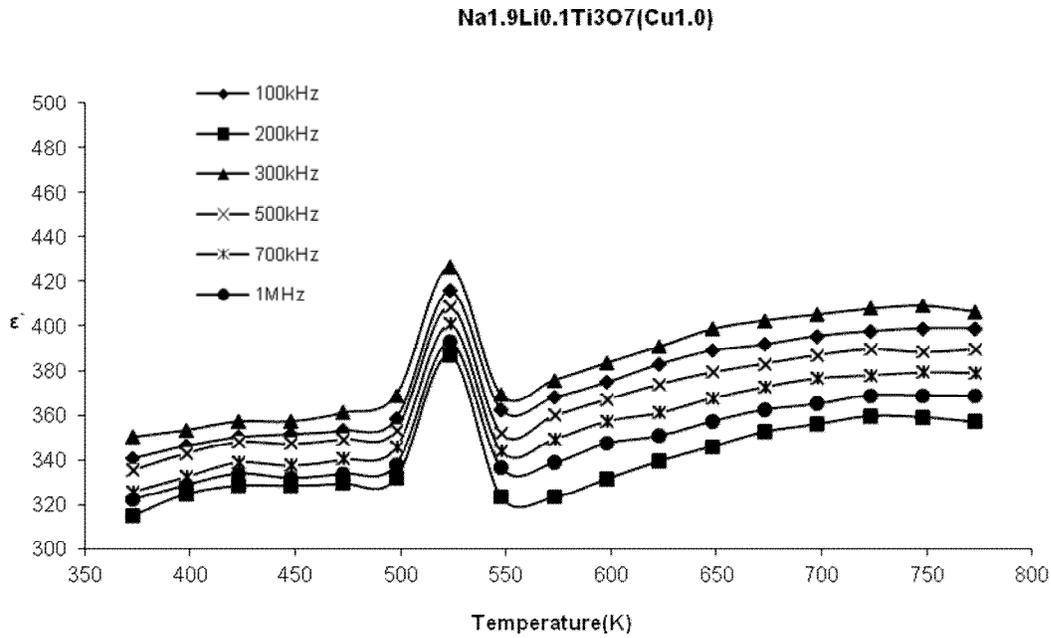


Figure 3(c). Dielectric constant ( $\epsilon'$ ) versus temperature for CSLT-2

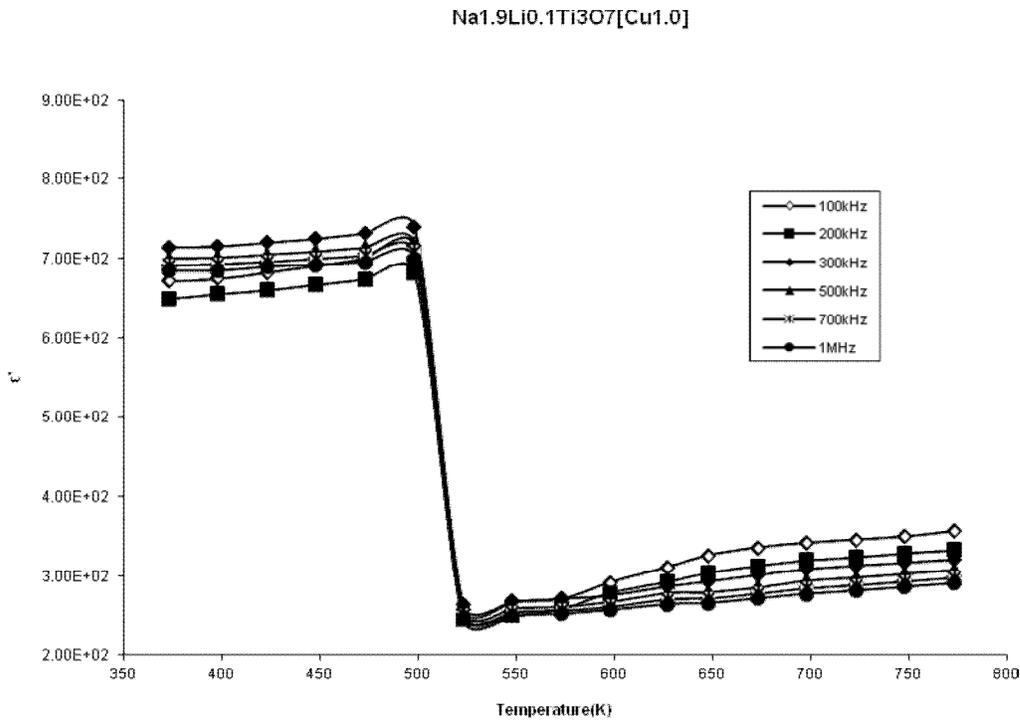
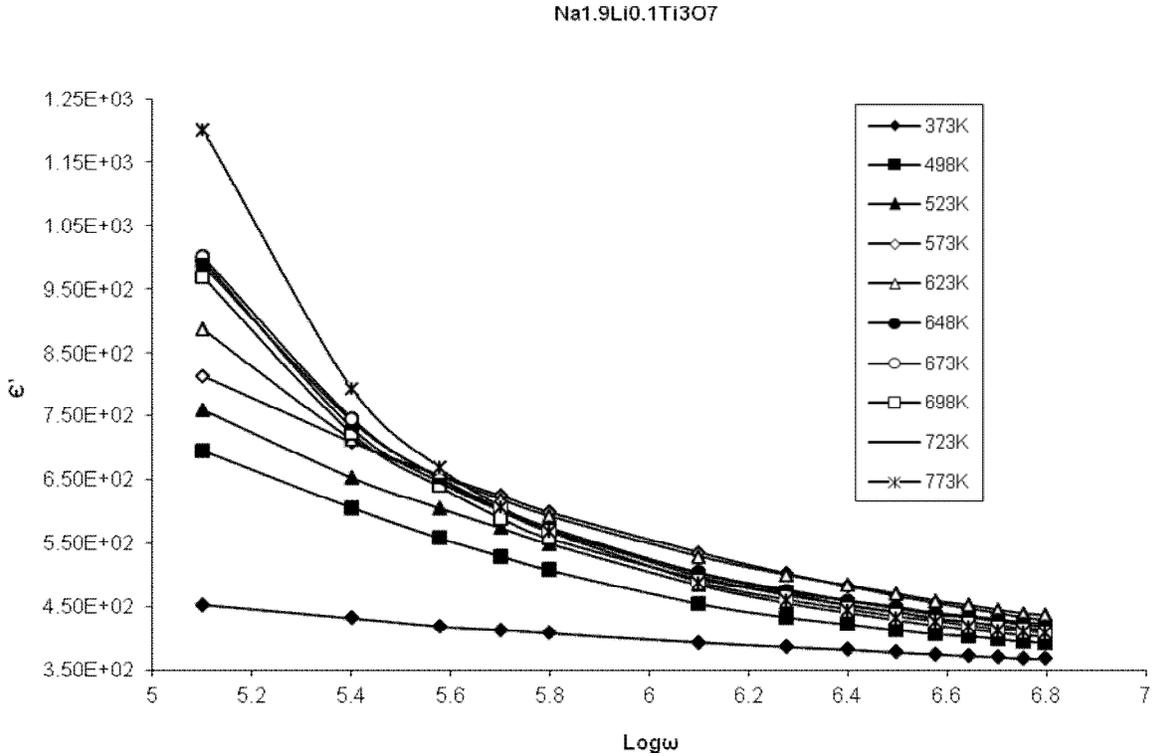
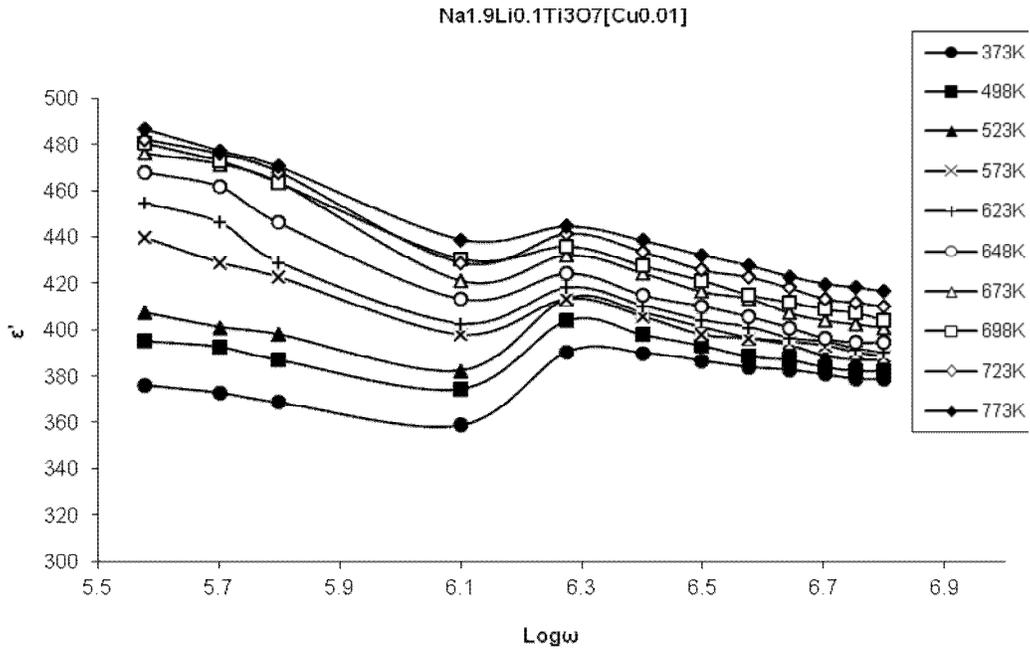


Figure 3 (d). Dielectric constant ( $\epsilon'$ ) versus temperature for CSLT-3

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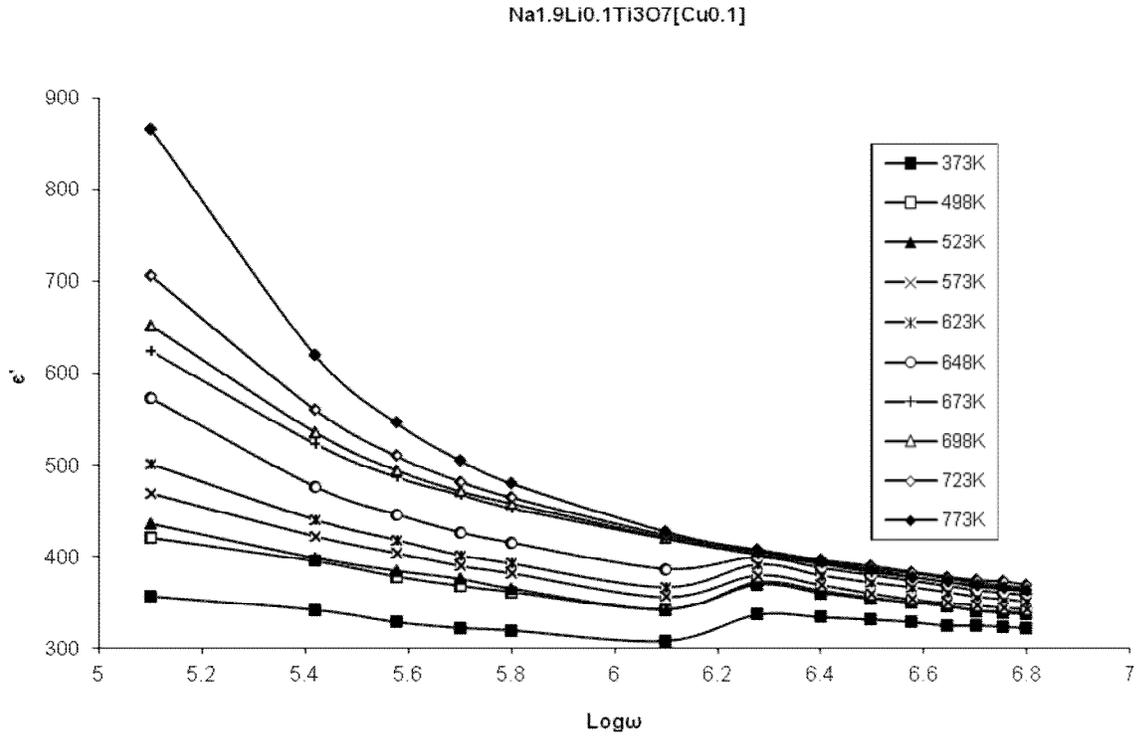


**Figure 4(a). Dielectric constant ( $\epsilon'$ ) versus frequency for SLT**

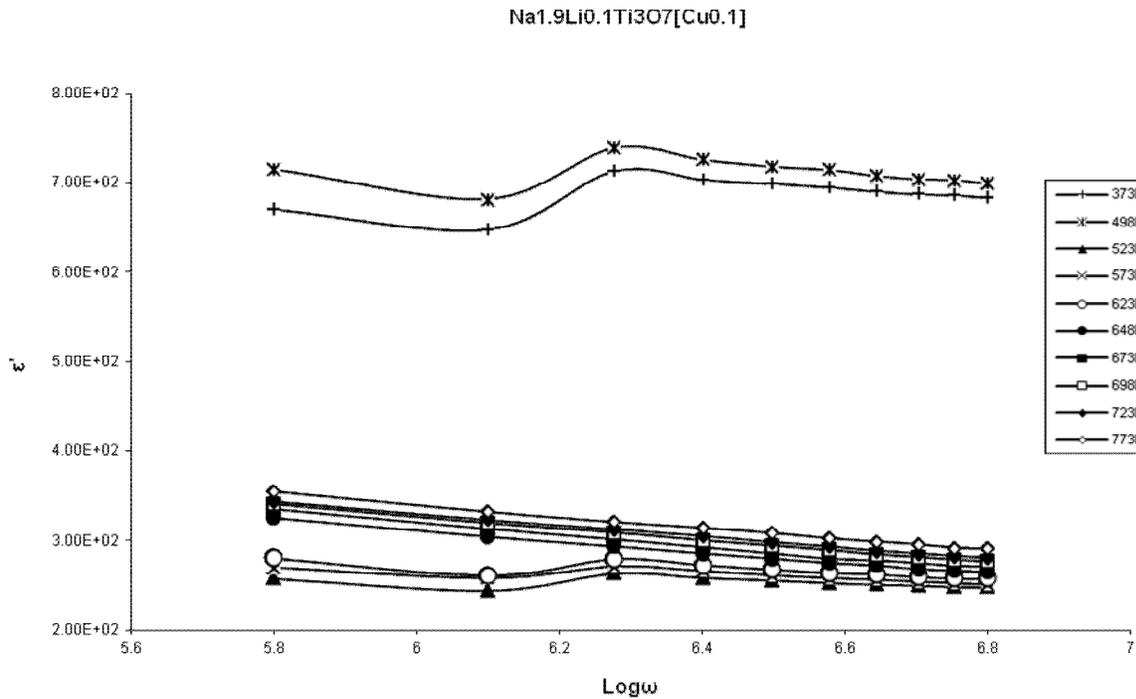


**Figure 4(b). Dielectric constant ( $\epsilon'$ ) versus frequency for CSLT-1**

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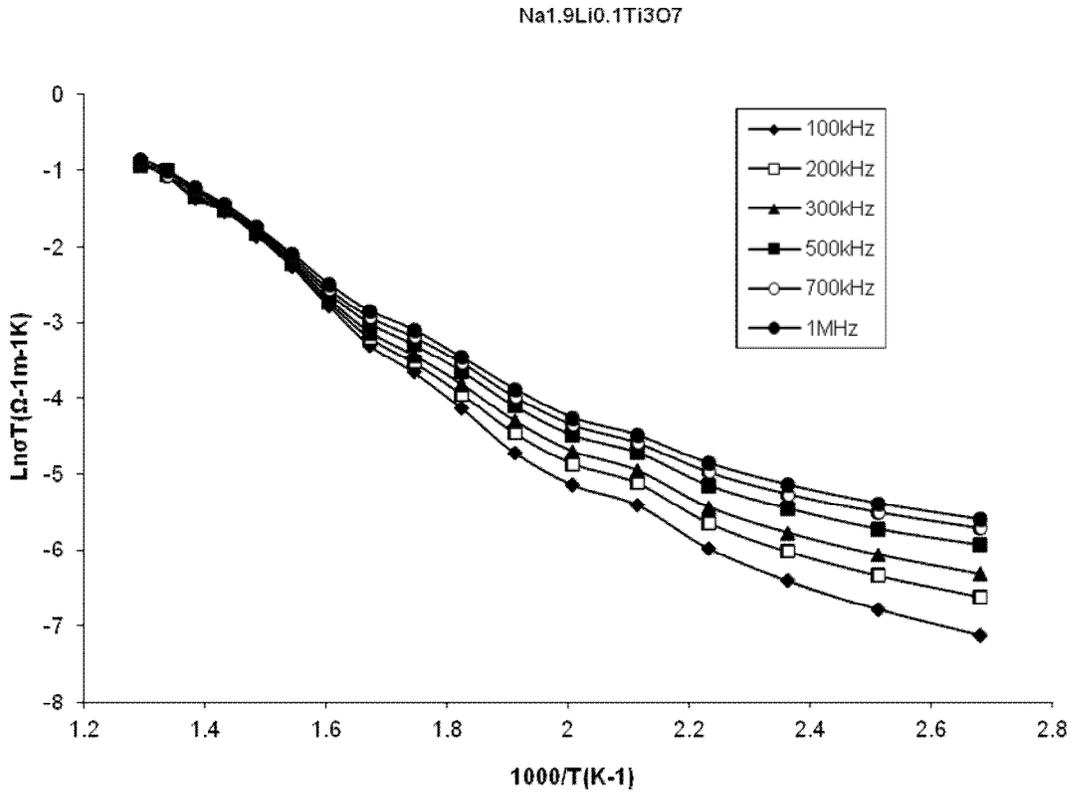


**Figure 4(c). Dielectric constant ( $\epsilon'$ ) versus frequency for CSLT-2**

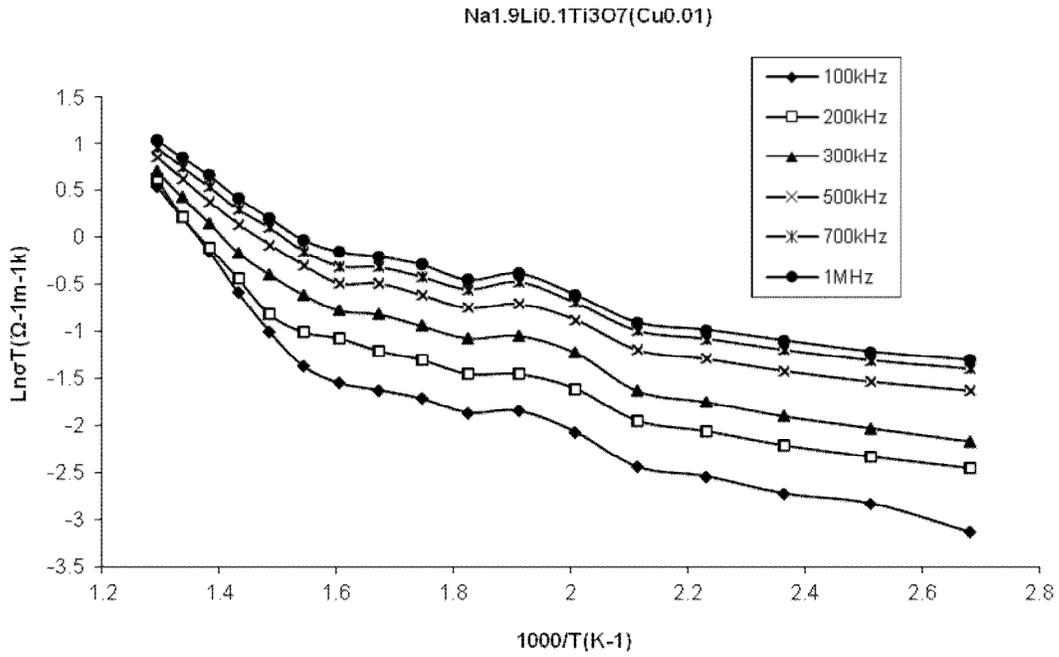


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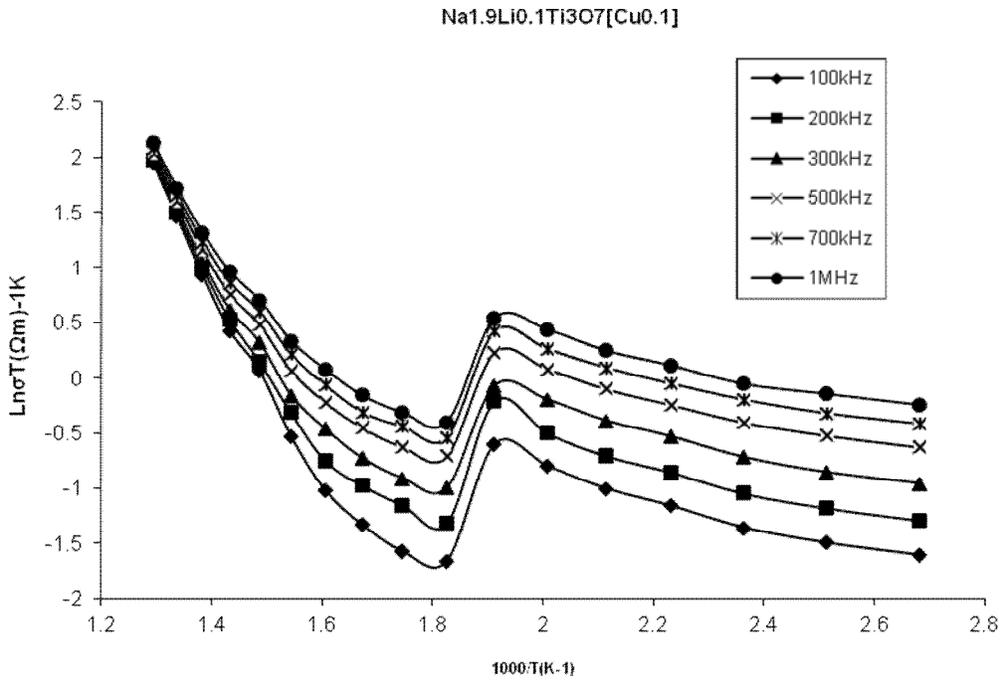
**Figure 4(d). Dielectric constant ( $\epsilon'$ ) versus frequency for CSLT-3**



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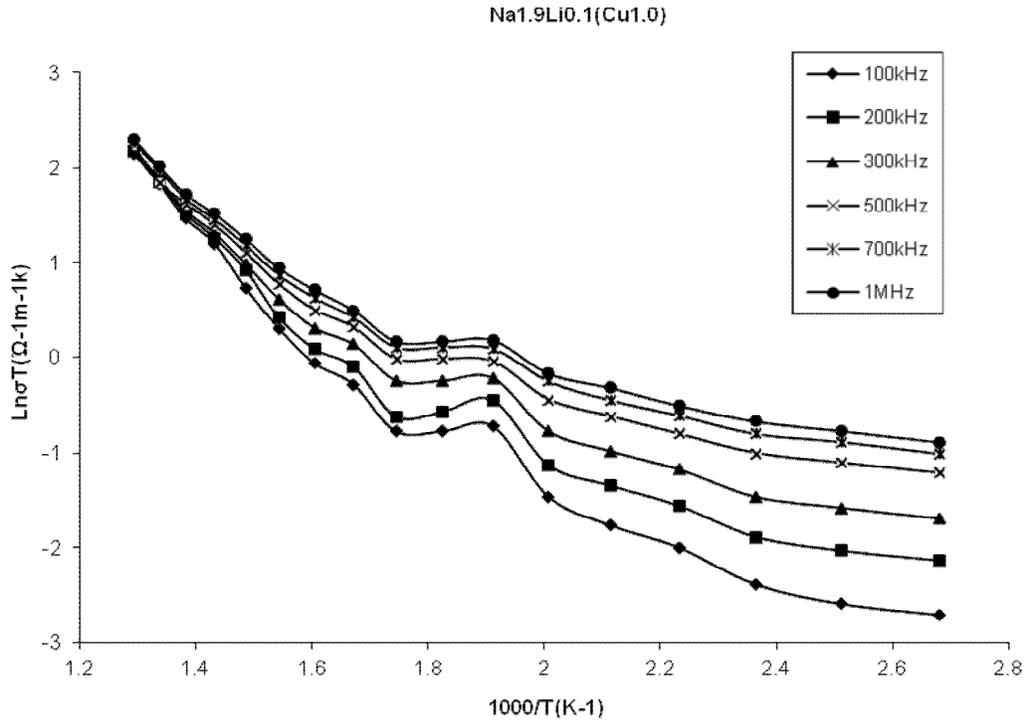


**Figure 5(b). LnσT versus 1000/T for CSLT-1**

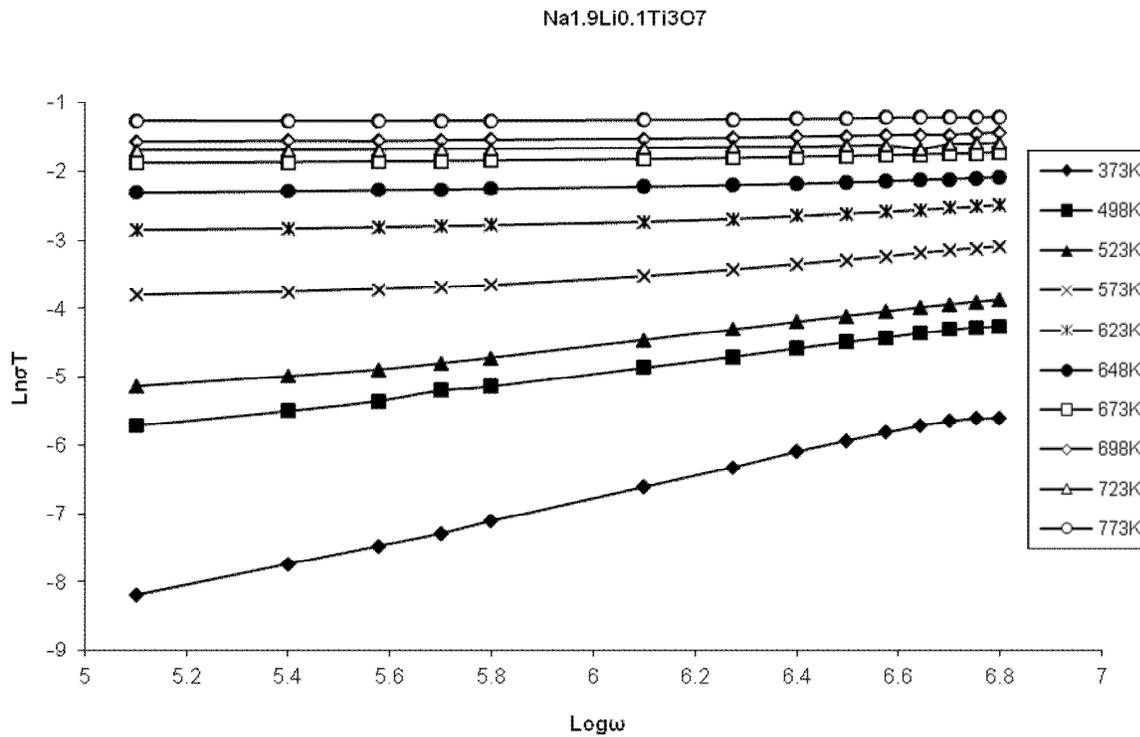


**Figure 5(c). LnσT versus 1000/T for CSLT-2**

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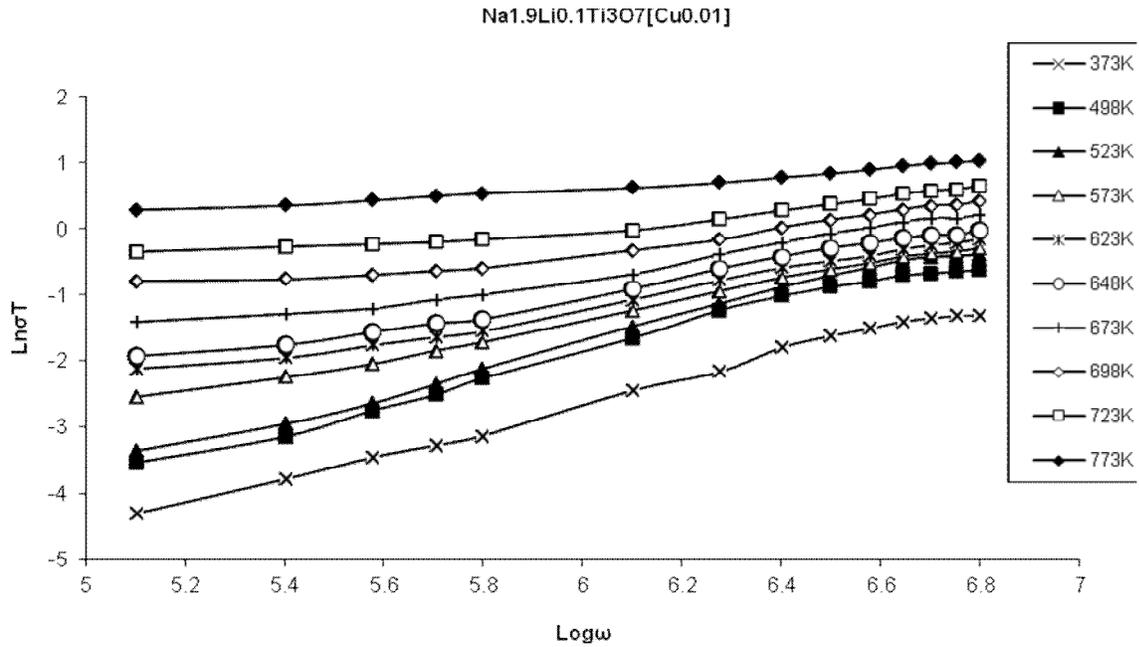


**Figure 5(d).  $\text{Ln}\sigma T$  versus  $1000/T$  for CSLT-3**

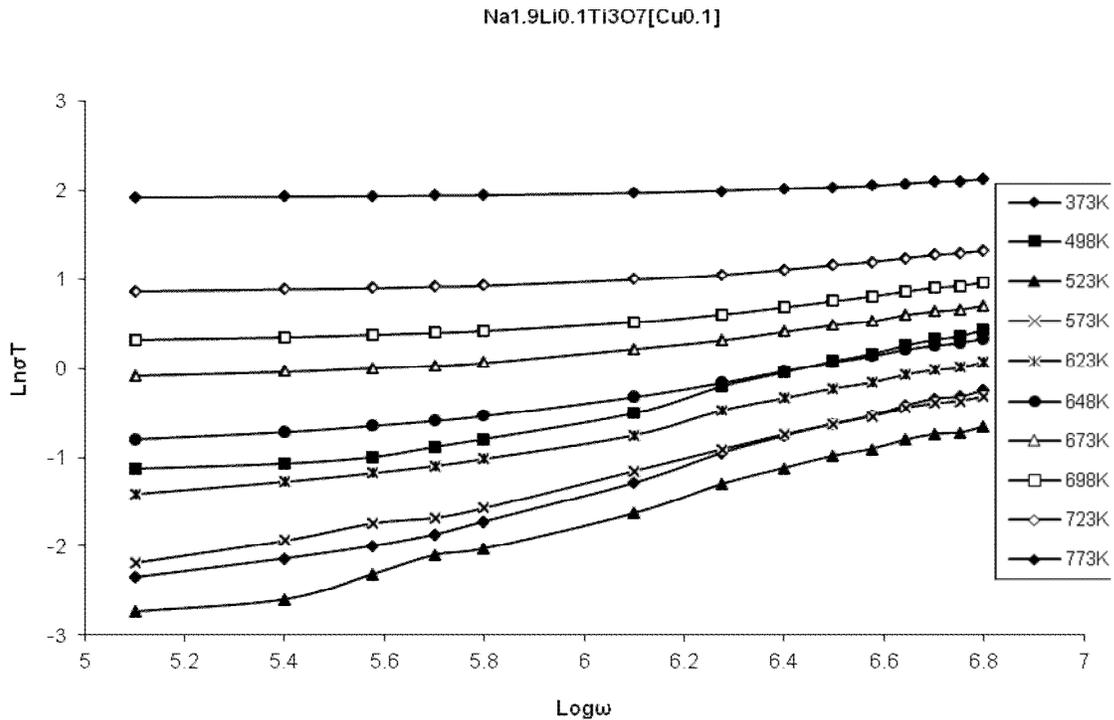


**Figure 6(a).  $\text{Ln}\sigma T$  versus frequency for SLT**

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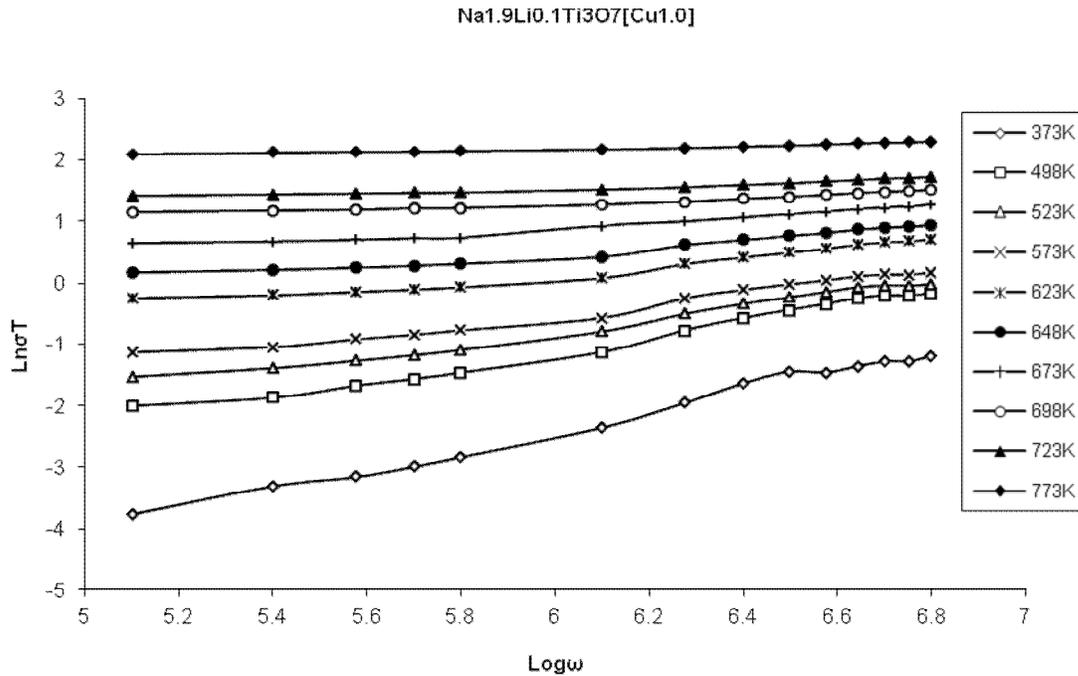


**Figure 6(b). LnσT versus frequency for CSLT-1**



**Figure 6(c). LnσT versus frequency for CSLT-2**

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**Figure 6(d).  $Lntan\delta$  versus frequency for CSLT-3**

Accordingly, the variation of  $\tan\delta$  with respect to temperature can be explained by assuming that the losses are of mixed type i.e. dipole losses and losses due to electrical conduction.

Fig. 2(a), (b), (c), (d) show the variation of  $\tan\delta$  with respect to frequency in temperature range 373-773K for SLT, CSLT-1, CSLT-2 and CSLT-3 respectively. The result shows that at lower temperature  $\tan\delta$  decreases gradually when the frequency increases but at higher temperature loss tangent ( $\tan\delta$ ) decreases exponentially with rise in frequency. This may be an indication of dipole mechanism of losses [31] along with the losses due to motion loosely bound ions, i. e. losses due to electrical conduction.[31]

Fig.3 (a), (b), (c), (d) show the variation of dielectric constant ( $\epsilon'$ ) with respect to temperature for SLT, CSLT-1, CSLT-2 and CSLT-3 respectively. The trend of variation with a broad peak at 598K for SLT and 523K for CSLT-1, and CSLT-2 is almost similar for pure and doped samples. The peak observed at 598K for SLT and 523K for CSLT-1, and CSLT-2 might be attributed to possible ferroelectric phase transition. The shifting of peak towards 523K for doped samples may be due to roles of dipoles generated due to the substitution of  $Cu^{2+}$  at  $Ti^{4+}$  sites. The remaining variation of all curves can easily be understood by proposing that dipoles are not aligned in low temperature region, so when temperature raises the orientation of dipoles is facilitated and this increases the dielectric constant. As the temperature grows the chaotic thermal oscillation of molecules is identified and degree of orderliness of their orientation is diminished.

For CSLT-3 the variation of dielectric constant ( $\epsilon'$ ) with rise in temperature at some fixed frequencies shows that the dielectric constant is strongly dependent on particular temperature. The dielectric constant ( $\epsilon'$ ) almost remains constant up to the temperature 523K and shows two times decrease in the temperature range 523K-548K. This corresponds to the Structural ferroelectrics phase transition of the material with transition temperature 523K.[32,33] After 548K the value of dielectric constant ( $\epsilon'$ ) again almost remains invariant up to 773K.

The dielectric constant ( $\epsilon'$ ) versus frequency curves are shown in fig.4 (a), (b), (c), (d) The curves show the same nature of variation as for polar dielectrics for SLT, CSLT-1 and CSLT-2 but entirely different

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nature for CSLT-3. The value of dielectric constant ( $\epsilon'$ ) is lower at higher frequency. On copper doping the value of dielectric constant ( $\epsilon'$ ) decreases. It is reported [Lingwal 2003] that when the frequency of alternating voltage increases the value of dielectric constant of polar dielectric remains invariable but beginning with a certain frequency ( $f_0$ ) when polarization fails to settle itself during one half period, dielectric constant ( $\epsilon'$ ) begins to drop approaching to very high frequencies the values to nonpolar dielectrics. Dielectric dispersion is seen for these compounds.

For CSLT-3, the value of dielectric constant ( $\epsilon'$ ) decreases up to 498K. After 498K it suddenly decreases and attains the value approximately two times lower than the value at 498K. After 523K the value of dielectric constant ( $\epsilon'$ ) increases very slowly up to the temperature range of study. This corresponds to the ferroelectric phase transition of the material with transition temperature 523K. [Kumar v2005, Lingwal 2003]

Fig.5 (a), (b), (c), (d) show the variation of conductivity curves  $\text{Ln}\sigma T$  versus  $1000/T$ . The obtained conductivity curves are divided into three regions for SLT, two regions for CSLT-1 and four regions for CSLT-2 and CSLT-3. The region wise discussion is given below.

**Region-I:** The almost frequency dependent and temperature independent region-I exists up to 548K for SLT with one small peak at 473K, 473K for CSLT-1, 473K for CSLT-2 and 523K for CSLT-3. The nature of a.c. conductivity can be interpreted by proposing that electronic hopping conduction plays major role in this region with lowest activation energy as already proposed. [Pal D. 2005] Many research groups have been interpreting such results by proposing that a.c. conductivity due to electronic hopping conduction increases with the frequency ( $\omega$ ) and is proportional to  $\omega^s$ , where  $s$  is a parameter having value less than unity. Such a frequency dependence, which has been attributed to a wide distribution of relaxation times due to distribution of jump distance and the barrier heights has been observed in a wide variety of low mobility materials. [Joncher 1972] Higher value of a. c. conductivity than d. c. conductivity reported by Shripal et al. may be explained by proposing that the exchangeable interlayer ionic and electronic hopping conduction co-exist in this temperature region. The appearance of peak at 473K in conductivity curve for SLT may be due to the presence of  $\text{Li}^+$  in the interlayer space results such type of Na-Li-Ti configuration, which appreciably reduces the number of loose electrons from  $\text{Ti}_3\text{O}_7^{2-}$  groups and subsequently suppresses the electronic hopping (polaron) conduction. However, for copper doped derivatives this interlayer ionic conduction disappears.

**Anomalous region:** This region exists from 473K to 648K for CSLT-1, from 473K to 573K for CSLT-2 and from 523K to 623K for CSLT-3. The existence of first peak at 523K for CSLT-1, CSLT-2 and CSLT-3 in conductivity curves may be due to microstructural phase change such a peak also appears in dielectric constant versus temperature plots at 523K for all copper doped derivatives. Furthermore, the observed peak at 598K in conductivity curve for CSLT-1, CSLT-2 and CSLT-3 may be due to substitution of  $\text{Cu}^{2+}$  at  $\text{Ti}^{4+}$  sites. Their substitution would naturally cause the number of loose electrons to increase appreciably to jump through Ti-Ti chains in conduction.

**Region II:** The almost temperature dependent and less frequency dependent region exist up to 648K for SLT, up to the temperature range of study for CSLT-1, from 573K to 698K for CSLT-2 and from 623K to 698K for CSLT-3. This can be explained by assuming that the interlayer ionic conduction dominates over electronic hopping conduction The slope of conductivity plots is higher than region I so the mechanism of conduction may be attributed due to associated interlayer ionic conduction as proposed by Shripal et al. [Shripal 1989]

**Region III:** The almost temperature dependent and frequency independent region III exists from 648K for SLT, from 698K for CSLT-2 and CSLT-3. The slope of conductivity curves for SLT, CSLT-2 and CSLT-3 are greater than those of region II. It seems that  $\text{Cu}^{2+}$  substitution at  $\text{Ti}^{4+}$  sites loosen off few oxygens, from  $\text{Ti}_3\text{O}_7^{2-}$  groups and then participate in conduction in this region. It seems that oxygens making dipoles with  $\text{Ti}^{4+}/\text{Cu}^{2+}$ , start to break at the lower limit of this region and then loosen off oxygens to participate in conduction. Thus, the conduction mechanism in this region may be understood as associated modified interlayer ionic conduction.

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Accordingly, the conduction mechanism in this region may be associated modified interlayer ionic conduction in which the loosen oxygens from  $\text{Ti}_3\text{O}_7^{2-}$  groups take part in conduction along with the associated interlayer alkali ions.

$\text{Ln}\sigma\text{T}$  versus frequency curves at different temperatures are shown in fig.6 (a),(b),(c), (d). From these curves it is clear that the dependence of a. c. conductivity decreases with increase in temperature. Thus the electronic hopping conduction is dominant at lowers temperatures and diminishes with the rise in temperatures.

### **CONCLUSIONS**

The outcomes of the present investigations may be summarized as:

1. Through dielectric-spectroscopic investigations the presence of dipole mechanism of losses along with the losses due motion of loosely bound ions and space charge polarization at higher temperatures have been identified
2. The possible ferroelectric phase transitions at 523K indicating the ferroelectric behavior for all the copper doped derivatives.
3. Two types of copper substitutions and dipoles in the interlayer space as well has been identified through  $\tan\delta$  versus temperature plots for all the doped derivatives.
4. Layered Sodium Lithium trititanates ( $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$ ) ceramic with 0.01,0.1 and 1.0 molar percentage of CuO doped derivatives can be put in the class of mixed ionic-electronic materials.

### **ACKNOWLEDGEMENTS**

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### **REFERENCES**

- Abraham and B. Bleany (1990)**. Electron Paramagnetic Resonance of Transition Ions (Clarendon Press Oxford)
- Anderson and Wadsley (1961)**. The crystal structure of  $\text{Na}_2\text{Ti}_3\text{O}_7$ . *Acta crystallography* **14**, 1245.
- Machida, Ma X.W. H, Taniguchi, Yabunka, and Kiyina (2000)**. Pillaring and photocatalytic property of partially substituted layered titanates,  $\text{Na}_2\text{Ti}_{3-x}\text{M}_x\text{O}_7$  and  $\text{K}_2\text{Ti}_{4-x}\text{M}_x\text{O}_9$  (M=Mn, Fe, Co, Ni, Cu). *Journal of Molecular Catalysis*. **155**, 131.
- Maurya, Kumar, Shripal (2005)**. Dielectric-spectroscopic and a.c. conductivity studies on layered  $\text{Na}_{2-x}\text{K}_x\text{Ti}_3\text{O}_7$  (X=0.2, 0.3, 0.4) ceramics, *Journal of Physics and Chemistry of solids* **66**, 1614
- Pal D. and Shripal, (2007)** EPR and mixed electronic-ionic conductivity studies of pure and manganese doped layered Potassium–Lithium tetra titanates ( $\text{K}_{1.9}\text{Li}_{0.1}\text{Ti}_4\text{O}_9$ ). *Journal of Material Sciences: Materials in Electronics* **18**(4), 401.
- Pal D., Premchand, Tandon, Shripal, (2005)**. EPR and electrical studies in layered  $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$  and its copper doped derivatives. *Journal of the Korean Chemical Society* **49**(6) 560.
- Pal Dharmendra and Pandey J.L. (2010)**. Effect of paramagnetic manganese ions doping on frequency and high temperature dielectric response of Layered  $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$  ceramics. *Bulletin of Material Sciences* **33**(6) 691.
- Pal Dharmendra, Pal R.K., Pandey, Abdi Agnihotri, (2010)**. Bulk ac conductivity studied of Lithium substituted layered Sodium tri titanates ( $\text{Na}_2\text{Ti}_3\text{O}_7$ ). *Journal of Material Sciences: Materials in Electronics*. **21**, 1181.
- Pal Dharmendra, Pandey J.L., Shripal, (2009)**. Dielectric spectroscopic and ac conductivity investigations on manganese doped layered  $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$  ceramics. *Journal of the Korean Chemical Society* **53**(1), 42.

**Research Article**

**Pal Dharmendra, Prem Chand and Shripal (2004)** Preparation and characterization of Pure and manganese doped Layered  $\text{Na}_{1.9}\text{Li}_{0.1}\text{Ti}_3\text{O}_7$ , Proceeding of NSFD-XIII, 23-25.

**Shripal, Badhwar Maurya, Kumar and Tandon (2005)**. Dielectric and spectroscopic investigations, Advances in Condensed Matter Physics, edited by K.K. Raina (Allied Publisher New Delhi,) 250.

**Shripal, Badhwar, Maurya, Kumar, and Tandon, (2005)**. Dielectric and a.c. conductivity studies in pure and manganese doped layered  $\text{K}_2\text{Ti}_4\text{O}_9$  ceramics. *Journal of Material Sciences: Materials in Electronics*. **16**, 495

**Shripal, Mishra, and Pandey, and Tandon (1992)**. Electrical conductivity and epr investigations in iron doped polycrystalline  $\text{K}_2\text{Ti}_4\text{O}_9$ . *European Journal of Solid State Inorganic Chemistry*. **29**. **229**.

**Shripal, Pandey S.D., and Premchand, (1989)**. Electrical studies in some alkali titanates, *Solid State Communications*. **69**, 1203

**Shripal, Premchand and Pandey (1991)**. Electrical conductivity and electron paramagnetic resonance investigations in manganese-doped polycrystalline  $\text{Na}_2\text{Ti}_3\text{O}_7$ . *Journal of Material Sciences: Materials in Electronics*. **2** 89.

**Shripal, Tandon, and Pandey (1991)** Electrical conductivity and epr investigations in iron doped polycrystalline  $\text{K}_2\text{Ti}_4\text{O}_9$ . *Journal of Physics and Chemistry of Solids*. **52**, 1101.

**Tareev B. (1979)**. Physics of Dielectric Materials, (Mir Publishers, Moscow).

**Yakubovich and Kireev (2003)**. Refinement of Crystal structure of  $\text{Na}_2\text{Ti}_3\text{O}_7$ , *Crystallography Report*. **48**, 24.