

Research Article

A.C. Conductivity Investigations on Layered $\text{Na}_{2-x-y}\text{Li}_x\text{K}_y\text{Ti}_3\text{O}_7$ Ceramics

Rakesh Singh and Shripal

Department of Physics, P.P.N. College, Kanpur 208001, India

Correspondence should be addressed to Shripal; shripalsharmappn@gmail.com

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Frequency and temperature dependence of a.c. electrical conductivity of layered mixed ionic alkali trititanates, $\text{Na}_{1.89}\text{Li}_{0.10}\text{K}_{0.01}\text{Ti}_3\text{O}_7$, $\text{Na}_{1.88}\text{Li}_{0.10}\text{K}_{0.02}\text{Ti}_3\text{O}_7$, $\text{Na}_{1.86}\text{Li}_{0.10}\text{K}_{0.04}\text{Ti}_3\text{O}_7$, and $\text{Na}_{1.85}\text{Li}_{0.10}\text{K}_{0.05}\text{Ti}_3\text{O}_7$, have been investigated over a wide temperature $350\text{ K} \leq T \leq 725\text{ K}$ and frequency 10 kHz to 1 MHz range. For this, Arrhenius plots are used for a.c. electrical conductivity of these compounds. The obtained conductivity plots have been divided into four distinct regions and discussed the relevant theory. According to slope variation, the conduction mechanisms occurring are different in different temperature regions. At lower temperatures, the hopping electron disorders the surroundings by moving to its neighboring Ti atoms from their equilibrium positions, causing structural defect in the polycrystalline network named small polaron. At higher temperatures, associated/unassociated interlayer ionic conduction occurs along with the alkali ions hopping through the interlayer space and electron hopping (small polaron) conduction through Ti–Ti chains in these layered polar alkali titanates.

1. Introduction

Sodium trititanate ($\text{Na}_2\text{Ti}_3\text{O}_7$) is a member of alkali metal titanates $\text{A}_2\text{Ti}_n\text{O}_{2n+1}$ (A = alkali metal and $2 \leq n \leq 9$) family [1–5] whose crystal structure was first reported by Anderson and Wadsley [4]. Yakubovich and Kireev [6] refined its crystal structure with monoclinic unit-cell dimensions $a = 9.1332(2)\text{ Å}$, $b = 3.806(1)\text{ Å}$, $c = 8.556(2)\text{ Å}$, $\beta = 101.57(3)^\circ$, and space group = $P2_1/m$. The basic framework of $\text{Na}_2\text{Ti}_3\text{O}_7$ is built up by three TiO_6 octahedra-sharing edges at one level, and these units are joined to similar blocks above and below by additional edge sharing that forms zigzag strings. These strings form a layered structure with the compositions of six $(\text{Ti}_3\text{O}_7)^{2-}$ like a two-dimensional sheet centered on the (100) plane. The interlayer spaces of layered inorganic solids are suitable for constructing well-organized molecular assemblies through intercalation [7, 8]. Layered titanates are gorgeous host materials for such purposes due to their semiconducting [9, 10] and ion exchange properties [11–16]. Investigations on photo-induced electron transfer between titanate sheets and rare earth ions [15] and methyl viologen [12] are also available. These layered structured titanates are a

kind of typical complex oxides and studied extensively as ion exchangers [17–20]. These materials are also used to fabricate porous materials and to fix radioactive metal ions [21, 22].

Moreover, Machida et al. [23] have reported pillaring and photo catalytic properties 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 and Cu). The results of a.c. conductivity and dielectric-spectroscopic studies of $\text{Na}_{2-x}\text{K}_x\text{Ti}_3\text{O}_7$ with $x = 0.2, 0.3, 0.4$ [24] can be viewed. In a recent work, various results of EPR, dielectric-spectroscopic and a.c. conductivity investigations on copper doped layered $\text{Na}_{1.7}\text{K}_{0.3}\text{Ti}_3\text{O}_7$ ceramics are reported [25]. Pyroelectric and d.c. conductivity investigations [26] on layered $\text{Na}_{2-x-y}\text{Li}_x\text{K}_y\text{Ti}_3\text{O}_7$ fine ceramics are also available in the literature. EPR and d.c. conductivity of manganese doped layered $\text{Na}_{1.86}\text{Li}_{0.10}\text{K}_{0.40}\text{Ti}_3\text{O}_7$ have been carried out and published [27].

The characterization of these layered $\text{Na}_{2-x-y}\text{Li}_x\text{K}_y\text{Ti}_3\text{O}_7$ ceramics only by dielectric-spectroscopic investigations [28] has been carried out. In the literature, no attempt has been made to understand the various conduction processes in these layered alkali titanate derivatives. Accordingly, it seems very important to characterize these ceramics through

a.c. conductivity measurements, and the corresponding results are interpreted in this paper.

2. Experimental

The ceramic samples of lithium and potassium substituted layered perovskite type $\text{Na}_2\text{Ti}_3\text{O}_7$, that is, $\text{Na}_{2-x-y}\text{Li}_x\text{K}_y\text{Ti}_3\text{O}_7$ with $x = 0.10$, $y = 0.01, 0.02, 0.04$, and 0.05 , were prepared by conventional sintering process using high purity chemicals and described elsewhere [28]. Thus, the compounds included in this work are $\text{Na}_{1.89}\text{Li}_{0.10}\text{K}_{0.01}\text{Ti}_3\text{O}_7$, $\text{Na}_{1.88}\text{Li}_{0.10}\text{K}_{0.02}\text{Ti}_3\text{O}_7$, $\text{Na}_{1.86}\text{Li}_{0.10}\text{K}_{0.04}\text{Ti}_3\text{O}_7$ and $\text{Na}_{1.85}\text{Li}_{0.10}\text{K}_{0.05}\text{Ti}_3\text{O}_7$, and symbolized as SLPT-1, SLPT-2, SLPT-3, and SLPT-4, respectively.

For a.c. conductivity measurements, a pelletized sample has been mounted in a sample holder and introduced in a cylindrical furnace. The chamber containing the sample-holder is evacuated up to 10^{-3} mbar pressure. The upper part of the sample holder is kept cool with the continuous flow of chilled water. The capacitance (C_p), relative permittivity (ϵ_r), and the loss tangent ($\tan\delta$) have been measured directly using a precision digital LCR meter (Agilent 4284 A) in the frequency scan from 10 kHz to 1 MHz at a temperature rate of 3 K/minute from 350 K $< T < 725$ K with the help of a programmable temperature controller at the desired values.

3. Results and Discussion

Figures 1(a)–1(d), depict Arrhenius plots of a.c. electrical conductivity for the SLPT-1, SLPT-2, SLPT-3, and SLPT-4 mixed electronic-ionic layered polycrystalline ceramics, respectively. According to slope variation, the natures of conduction mechanisms occurring are different in different temperature regions. Accordingly, the obtained conductivity plots are divided into various distinct regions and may be discussed region wise as follows.

3.1. Region I. This strong frequency-dependent region, extends up to 500 K and 550 K for SLPT-1 (Figure 1(a)) and SLPT-2 (Figure 1(b)), respectively, but the region exists only up to 450 K for both SLPT-3 (Figure 1(c)) and SLPT-4 (Figure 1(d)) samples. It is observed that the behavior of a.c. electrical conductivity is very lightly temperature dependent for all the compositions in this lowest temperature region. As already proposed [29–31], this nature of a.c. conductivity can be interpreted by proposing that the electronic hopping conduction through Ti–Ti chains plays major role in this region. Many research groups [32] have already interpreted such results by proposing that a.c. conductivity due to electron hopping conduction increases as $A\omega^s$, where A is a constant dependent on temperature and s is the frequency exponent having values ≤ 1 and may be weakly temperature dependent. Such frequency dependence attributed to a wide distribution of relaxation times due to distribution of jump distances [33] and barrier heights [34] and has been observed in a wide variety of low-mobility materials [35]. Higher values of a.c. conductivity than those reported earlier [26] for d.c. conductivity investigation may be explained by knowing the

fact that the exchangeable interlayer ionic conduction coexist in this lowest temperature region.

Furthermore, the low-frequency dispersion is suggestive of the presence of accumulation of charges at the material-electrode interface (interfacial or space charge polarization as already detected in dielectric-spectroscopic investigations [28]) reflecting a drop in conductivity. The indication of saturation towards higher frequencies may be due to the release of space charge leading to enhancement in the conductivity. However, very strong frequency dependence in this region is due to a nonexponential type conductivity relaxation phenomenon, which may be a typical feature of an exchangeable interlayer ionic conduction (hopping process). More specifically, as frequency increases, a.c. conductivity increases due to the strong mobility of electrons hopping through TiO_6 - TiO_6 octahedral ribbons [29–31] and exchangeable interlayer alkali ions which are responsible for hopping mechanism of ionic conduction as well.

Enhancement in the upper limit of this region for SLPT-2 (550 K) and decrement again for SLPT-3 and SLPT-4 (450 K) can be explained by proposing that the K^+ ions initially substitute at $\text{Na}_{(2)}$ with seven-fold coordination sites [4] up to SLPT-2 and then replace $\text{Na}_{(1)}$ sites [4] with nine-fold coordinations with oxygen in the interlayer space, as each $\text{Na}_{(1)}$ is loosely situated than $\text{Na}_{(2)}$ in the interlayer space.

Accordingly, the conduction mechanism in this lower temperature region may be understood as follows: electronic hopping conduction is quite probable and could occur through Ti–Ti chains along with exchangeable (alkali ion hopping) interlayer ionic conduction through interlayer space existing in these layered materials.

3.2. Region II. In this region, the nature of a.c. conductivity is both frequent (slightly lesser) and maximum temperature dependent giving maximum slope. This region exists in the temperature range of 500–600 K and 550–650 K for SLPT-1 and SLPT-2 samples, respectively, but this region extends in the temperature range of 450–600 K for both SLPT-3 and SLPT-4 compounds. From Figures 1(a)–1(d), it can be observed that this region is further sub-divided as IIa and IIb, respectively. The subregion IIa can be seen from 500 to 550 K, 550 to 600 K, 450 to 500 K, and 450 to 500 K for SLPT-1, SLPT-2, SLPT-3, and SLPT-4 samples, respectively. The remaining span of temperature range is marked as the subregion IIb. The occurrence of IIb region for all the compositions (especially above 100 kHz frequencies of applied a.c. signal) is an outcome of space charge polarization which becomes dominant mechanism (up to the upper limit of region IIb) in transport process that is absent at frequencies ≥ 100 kHz.

Enhancement of the temperature range for SLPT-2 and decrement of the temperature range for SLPT-3 and SLPT-4 can easily be understood by the fact of substitution of K^+ at $\text{Na}_{(1)}$ and $\text{Na}_{(2)}$ sites as proposed which discussing the results of region I above. This is due to electron hopping through Ti–Ti chains of $(\text{Ti}_3\text{O}_7)^{2-}$ two-dimensional layers and alkali ion hopping process occurring through alkali ion sites in interlayer space existing between the opposite $(\text{Ti}_3\text{O}_7)^{2-}$ layers, respectively. The ionic conductivity involves associated

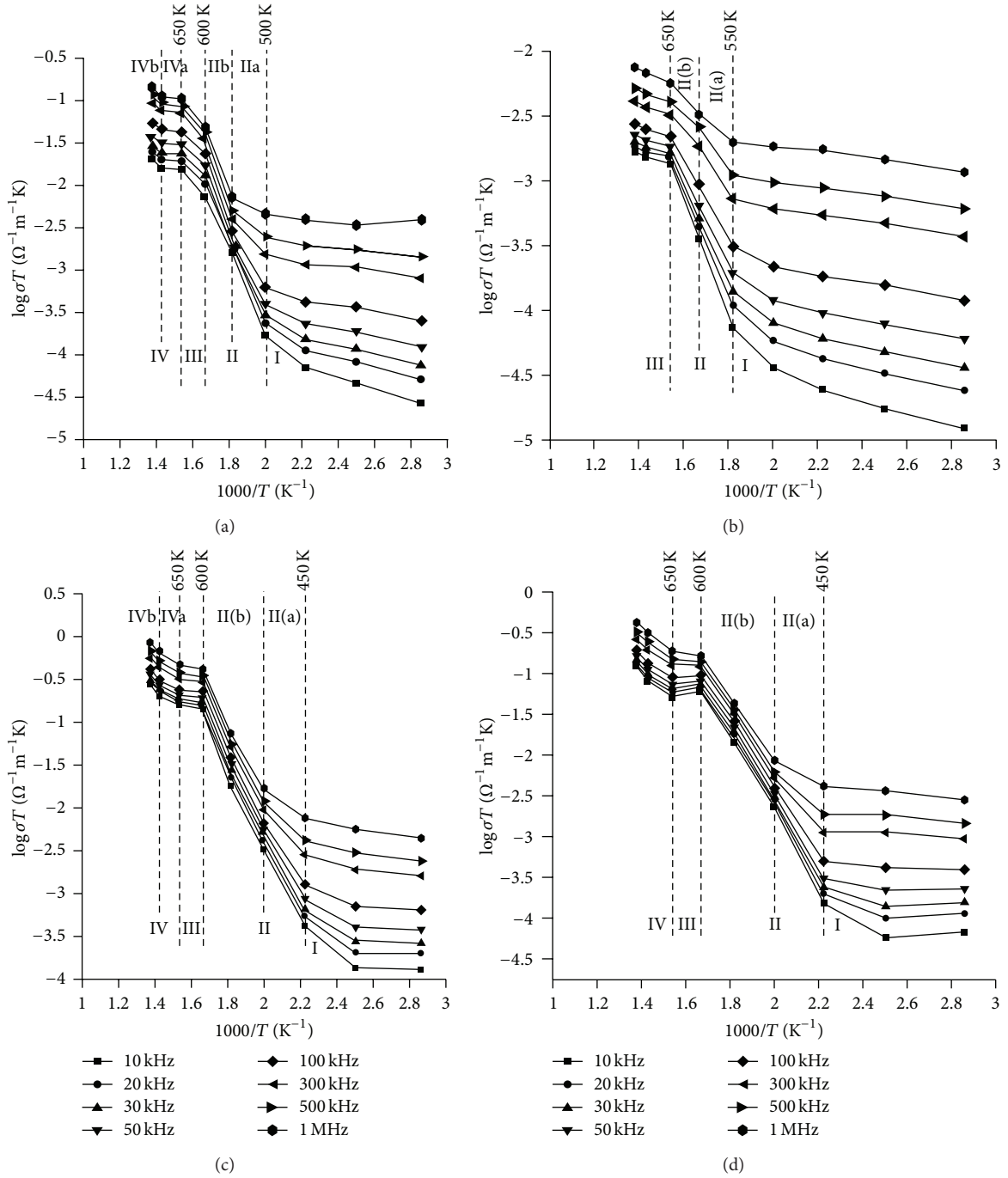


FIGURE 1: Log (σT) versus $1000/T$ plots at different frequencies for (a) SLPT-1, (b) SLPT-2, (c) SLPT-3, and (d) SLPT-4.

interlayer ionic conduction, unassociated interlayer ionic conduction, and modified interlayer ionic conduction as proposed earlier [36], whichever occurs and dominates over this region and in the next higher temperature regions. In addition, such a contribution includes the role of different dipoles as discussed by Shripal et al. [28] in a very recent communication and is responsible for giving a trend of variations in $\tan \delta$ and permittivity (ϵ) values with temperature and frequency.

Accordingly, the conduction mechanism in this temperature region can be understood as associated interlayer ionic conduction along with the alkali ion hopping through the interlayer space and electron hopping (small polaron) conduction through Ti-Ti chains in these layered polar alkali titanates.

3.3. Region III. This region exists from 600 to 650 K for SLPT-1, SLPT-3, and SLPT-4 samples, but it extends from

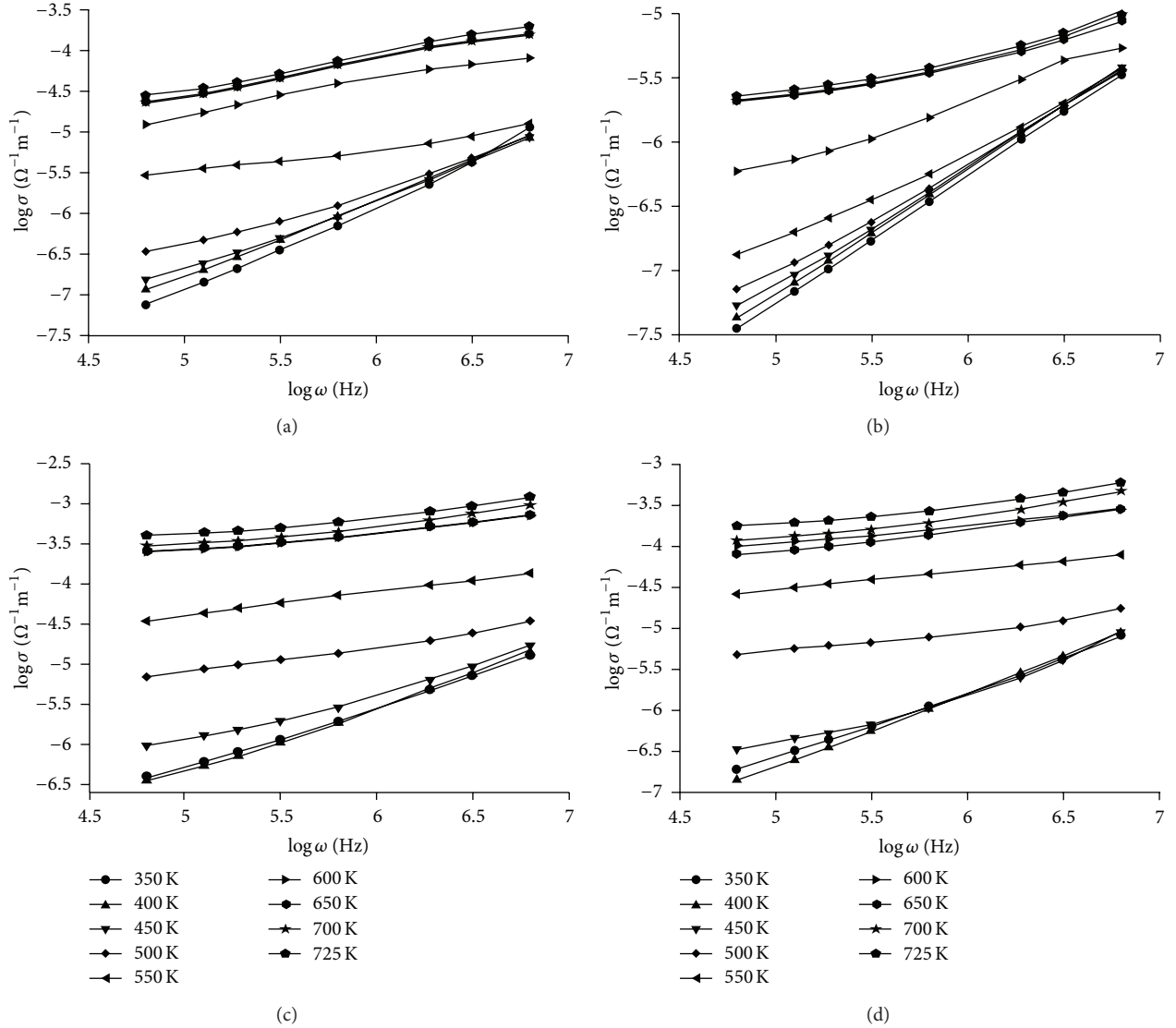


FIGURE 2: Log (σ) versus frequency plots at different temperatures for (a) SLPT-1, (b) SLPT-2, (c) SLPT-3 and (d) SLPT-4.

650 K to the temperature range of study for SLPT-2 sample. The slopes in this temperature region are lesser than the slopes obtained in the region II for SLPT-1 (Figure 1(a)) and SLPT-2 (Figure 1(b)) compositions. However, conductivity plots depict nearly temperature-independent conductivity for both SLPT-3 (Figure 1(c)) and SLPT-4 (Figure 1(d)) compositions in the plot. It is also observed that frequency dependence persists up to this higher temperature region as well.

However, I-V dipoles present in the interlayer space break up completely up to the upper limit of previous region II, and the mechanism of conduction can now be proposed as “unassociated interlayer ionic conduction” for SLPT-1 and SLPT-2 materials as proposed [36]. So, the two mechanisms involve unassociated interlayer ionic conduction with alkali ion hopping and operate for SLPT-1 and SLPT-2 compositions. For SLPT-3 and SLPT-4 materials, it seems that ionic conduction occurs through alkali ion

hopping (exchangeable) process, giving almost horizontal conductivity lines with the rise in temperature.

3.4. Region IV. This highest temperature region can be seen from 650 K to the temperature of study (725 K) for SLPT-1, SLPT-3, and SLPT-4 samples. However, the sample SLPT-2 does not show this region. This region can further be subdivided into IVa and IVb, respectively, for SLPT-1 and SLPT-3 samples. Appearance and explanation of region IVa for SLPT-1 seem to be similar to those of region III for SLPT-3 and SLPT-4 samples. However, this higher temperature region IVa bears mixed phenomenon, that is, mechanisms occurring in region III added by a partial unassociated interlayer ionic conduction mechanism of conduction in subregion IVb for SLPT-1 and SLPT-3 and in region IV for SLPT-4 are alkali ion hopping conduction and unassociated interlayer ionic conduction.

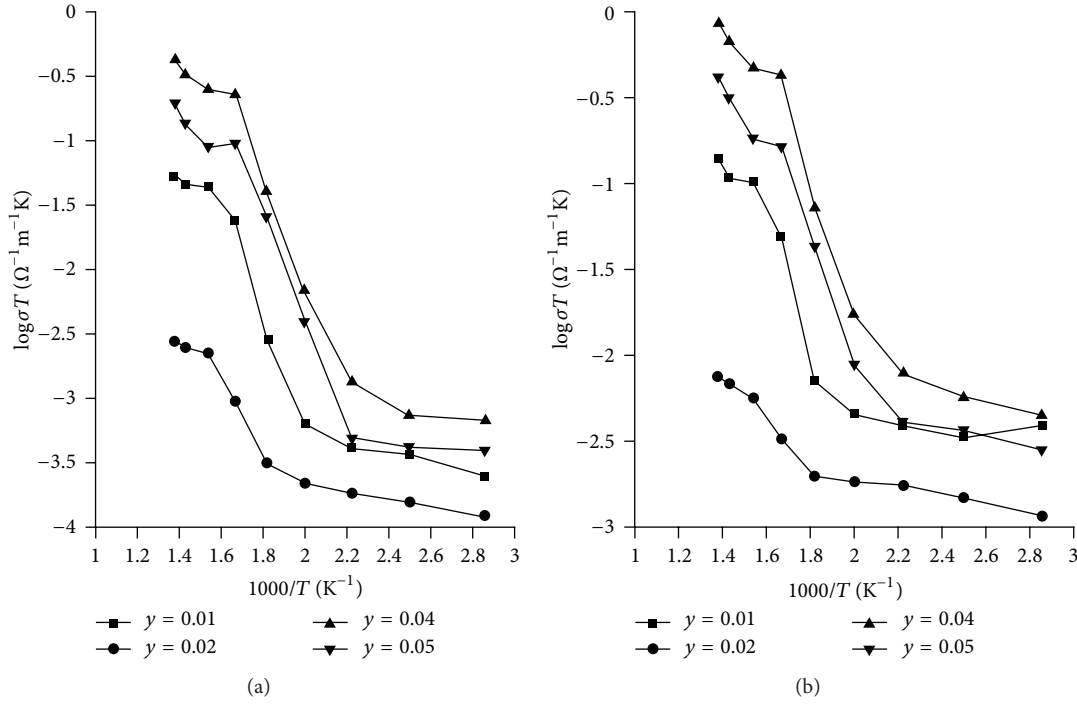


FIGURE 3: Log (σT) versus $1000/T$ plots at different K content bearing samples for (a) 100 kHz and (b) 1 MHz, respectively.

3.5. Log (σ) versus Frequency Plots. Log (σ) versus frequency plots for the samples SLPT-1, SLPT-2, SLPT-3, and SLPT-4 is shown in Figures 2(a)–2(d), respectively. From these curves it is observed that the dependence of conductivity on frequency decreases with the rise in temperature for all these compositions. Thus, electron hopping (polaron) conduction decreases with rise in temperature.

3.6. Conductivity versus K Content Plots. Figures 3(a) and 3(b) show a comparison of the conductivity values of different K content bearing samples at 100 kHz and 1 MHz, respectively. Interestingly, the conductivity values decrease to the least for SLPT-2, increases for SLPT-3, and decreases again for SLPT-4 at all temperatures. This can easily be understood by K^+ substitutions at two different $\text{Na}_{(1)}$ and $\text{Na}_{(2)}$ sites in the interlayer space and discussed as previously (region II). The significant result is that the decrement of electrical conductivity is an outcome of decreased ion migration due to replacement of large and heavy K^+ (ionic radius 1.33 Å) and small and light Li^+ (ionic radius 0.60 Å) at some of the medium Na^+ (ionic radius 0.95 Å) ions at both types of $\text{Na}_{(1)}$ and $\text{Na}_{(2)}$ sites. Accordingly, the creation of microstructural heterogeneity in the interlayer space along with the TiO_6 octahedral ribbons can be proposed.

4. Conclusions

In sum, the present study reveals the following points. (i) For the first time, we have characterized the lithium and potassium mixed $\text{Na}_2\text{Ti}_3\text{O}_7$ layered polycrystalline (SLPT-1, SLPT-2, SLPT-3, and SLPT-4) through a.c. electrical conductivity investigations. (ii) We have proposed that the K^+

ions initially substitute $\text{Na}_{(2)}$ with seven-fold coordination sites and then replace $\text{Na}_{(1)}$ sites with nine-fold coordinations with oxygen in the interlayer space. (iii) We have found very strong frequency dependence of conductivity in lower temperature region that may be due to a nonexponential type conductivity relaxation phenomenon reflecting a typical feature of an exchangeable interlayer ionic conduction. (iv) Finally, the decrement in electrical conductivity is an outcome of decreased ion migration due to replacement of large and heavy K^+ (ionic radius 1.33 Å) and small and light Li^+ (ionic radius 0.60 Å) at some of the medium Na^+ (ionic radius 0.95 Å) at both types of $\text{Na}_{(1)}$ and $\text{Na}_{(2)}$ sites and hence creation of microstructural heterogeneity in the interlayer space along with the TiO_6 octahedral ribbons. (v) These polycrystalline layered $\text{Na}_{2-x-y}\text{Li}_x\text{K}_y\text{Ti}_3\text{O}_7$ fine ceramics can be put in a class of mixed ionic-electronic materials. Accordingly, these materials can be investigated for the preparation, kinetics, and performance of mixed ionic- electronic conducting electrodes (MIEE) in favor of the application in an alkali metal for thermal to electric converter (AMTEC). Using these materials, the developing of an MIEE can be an attractive alternative to improve the efficiency of AMTEC.

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