

Thermal Conductivity and Thermoelectric Power of Potassium and Sodium-Substituted Bi-2212 Superconductor prepared by PEI Technique

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Abstract:

In the present study, thermal conductivity (κ - T), thermoelectric power (S - T), and figure of merit (ZT) measurements from room temperature down to 20 K of polycrystalline $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Na}_x\text{Cu}_2\text{O}_{8+y}$ and $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{K}_x\text{Cu}_2\text{O}_{8+y}$ superconductors prepared by a polymer solution technique using polyethyleneimine, PEI, have been carried out. The thermoelectric power, $S(T)$, of samples have both negative (at high T) and positive values (at low T), and are found to increase with decreasing temperature, reaching their maximum values (peaks) around their T_c values and dropping rapidly to zero below T_c . Na and K substitutions also have a considerable effect on the thermal conductivity, κ . Namely, with increasing Na concentration the magnitude of κ is decreased, but for the K substitution, the magnitude of κ is drastically increased for $x=0.75$.

Keywords: Bi-based cuprites; Transport properties; Electrical conductivity; Thermal conductivity; Thermoelectric power

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1. Introduction

Since Bi-based superconductors $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_x$ ($n = 1, 2, 3$) were discovered by Maeda *et al.* [1] many substitutions have been made in BSCCO system in order to improve their physical and magnetic properties [2-25]. High- T_C Bi-based superconductor family is characterized by a high superconducting transition temperature $T_C \sim 90$ and 110 K for the phases with $n = 2$, and 3 , respectively, and a high upper critical magnetic field, B_{c2} , which can reach 150 T [26]. The weak coupling between BiO–BiO layers in the BSCCO system enables substitution of rare-earth ions for Ca and/or Bi and makes the system interesting from the point of studying its structural, electrical, thermal and magnetic properties. It is well-known that the BSCCO system superconducting properties are very sensitive to the hole-carrier concentration which, in turn, depends on the substitution levels [27-30].

In general, the studies of transport properties, such as electrical resistivity, thermoelectric power (S) and thermal conductivity (κ), are important for exploring the conduction mechanism and the nature of the charge carriers. The thermoelectric power properties of BSCCO samples have been investigated by many research groups [31-39]. It was reported that S values have been found to be positive, negative or both, depending on the substituting element and proportion. In particular, a few studies related to substituting elements such as Fe, Li and Ti, for Cu in BSCCO system have exhibited positive S values around T_c and negative at temperatures well above T_c [32-35,40]. The presence of both positive and negative values of the thermoelectric power indicates the effect of two types of charge carriers, i.e. holes and electrons.

In addition, the material preparation technique plays a crucial role, as important as the substitutions in BSCCO systems. In order to easily obtain very homogeneous samples, the polymer matrix route is a very adequate solution method, as reported in previous works [41-44]. In this method, smaller particle sizes than the produced by the classical solid state method, are obtained leading to higher reactivity of powders and lower amount of secondary phases.

In order to analyze the thermoelectric power data, some theoretical models have been developed. Among these are: the Three State Model [45], the Hubbard-Hamiltonian model [46], the two-band model with Linear T-term [47], the Nagosa–Lee Model [48], the Phenomenological Narrow Band Model [49], and Xin' s two- band model [50]. However, no consensus has been reached among these models. There are also many studies on the thermal conductivity properties, $\kappa(T)$, of the *HTc* materials [51–56]. In these studies, it was generally observed that the thermal conductivity, κ , first decreases almost linearly with decreasing

temperature, then it suddenly increases below the T_c value, reaches a maximum value and then sharply drops to zero, at a temperature well below the relevant T_c . Two scenarios have been proposed to explain the rapid rise: Firstly, in the superconducting state ($T < T_c$) an increase in the carrier mean free path, due to a decrease of the phonon scattering causes the rapid rise in thermal conductivity [57,58]. Secondly, the electronic contribution in the Cu–O planes increases rapidly due to the strongly suppressed quasi-particle scattering rate in the superconducting state [59]. Recent experimental investigations, such as microwave conductivity and thermal Hall effect measurements, give an evidence for the increase in the electronic component around T_c [60,61].

2. Experimental Details

$\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Na}_x\text{Cu}_2\text{O}_y$ samples, with $x = 0, 0.05, 0.075, 0.1, 0.15, 0.20$ and $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{K}_x\text{Cu}_2\text{O}_y$ with $x = 0, 0.05, 0.075, 0.1$ have been prepared by using a polymer matrix route described in detail elsewhere [7,23]. In order to identify the present phases, powder X-ray diffraction patterns of the materials were recorded at room temperature using a Rigaku D/max-B powder diffractometer system working with $\text{CuK}\alpha$ radiation and a constant scan rate between $2\theta = 3$ and 60° . Resistivity and thermal characterization measurements were carried out in a Quantum Design PPMS system. The $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Na}_x\text{Cu}_2\text{O}_{8+y}$ samples, with $x = 0.05, 0.075, 0.1, 0.15,$ and 0.20 and the $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{K}_x\text{Cu}_2\text{O}_{8+y}$ samples, with $x = 0.05, 0.075,$ and 0.1 will be hereafter named as Na01, Na02, Na03, Na04, Na05, and K01, K02, K03, respectively.

3. Results And Discussion

3.1. XRD characterization

XRD patterns of pure, K and Na substituted samples are exhibited in Figs.1a and 1b, respectively. The main peaks correspond to the Bi-2212 phase in all cases (indicated by #2 in the graphs). It is clear that all samples are mainly composed by the Bi-2212 phase, independently of the K and Na amount. Moreover, small amounts of CaCuO_2 secondary phase (indicated by *) was detected in all the samples which is a clear indication that K and Na do not destabilize the Bi-2212 superconducting phase. The crystal symmetry of all samples was found as tetragonal structure and the calculated lattice parameters using the least squares method are tabulated in Table 1. As it can be inferred from these data, while c -parameter is decreasing, a - b parameters are slightly increasing when K or Na-content is increased. These

variations are due to the fact that the insertion of Na^{1+} substituting Ca^{2+} ions in the structure produces a decrease on the global positive charge in the structure. On the other hand, a modification on the oxygen content by substituting K or Na for Ca ions has to be produced to maintain the electrical neutrality in the structure, decreasing c -parameter and, consequently, raising the a - b . Additionally, it is found that no changes in crystal symmetry with variation of K or Na concentration have been observed.

Debye Scherrer formula [62] has been applied to the XRD data in order to get more information about the crystal sizes,

$$L_{hkl} = 0.9 \lambda / \beta \cos \theta \quad (1)$$

where λ is the used wavelength, β is the full width at half maximum of X-ray peaks and θ is the angle of the peak. As can be seen from Table 1, the particle sizes increase with increasing K or Na-concentration, probably due to the decrease of the system melting point induced by K or Na. As a consequence, the growth process speed is increased due to the formation of a small amount of liquid phase.

3.2. Charge Carrier Concentration

The calculation of the charge carrier concentration, p , in the CuO_2 planes is of fundamental interest to understand the mechanism of the doping dependence of transition temperature, T_c , in the HTc superconductors. Presland et al. [63] suggested the empirical relation;

$$T_c/T_c^{max} = 1 - 82.6 (p - 0.16)^2 \quad (2)$$

between T_c and Sr-hole doping in $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$, where T_c^{max} is the maximum attainable T_c , and p is the hole concentration per Cu atom in the CuO_2 planes. According to Eq.2, T_c is maximized for hole concentrations ≈ 0.16 and falls to zero at ≈ 0.05 on the underdoped side and at ≈ 0.27 on the overdoped side. The above relation was obtained to be a valuable model to find out the T_c values versus doping concentration [64]. Therefore, the relation has often been used as a convenient representation of the relation between T_c and the hole concentration for all the hole doped cuprates. The calculated p values for the samples are displayed in Table 1. These results show that the p -values are very close to each other. However, the values of samples with Na are close to the obtained in the undoped Bi-2223 phase. The findings indicate that the amount of Na dopant is far from the overdoping limit and for the samples with K is close to underdoped side.

3.3. Thermoelectric Power

The thermoelectric power, S , provides information concerning the nature of charge carriers, carrier concentration and band structure, since it is highly sensitive to the charge transport mechanism. Therefore, many experimental research on carrier concentration and temperature dependence of S have been performed for the Bi-based system [37,38,65]. As it is well known, the high- T_c superconducting oxides are strongly correlated systems. Based on the Hubbard model, the phenomenological expression of thermoelectric power at high temperatures was derived by different groups [66,67], and subsequently modified by Cooper et al. [68]. According to these studies, the thermoelectric power of HTc materials at around room temperature can be described by the formula;

$$S = k_B / e [\ln((1-p)/2p) - \ln 2] \quad (3)$$

where p , k_B and e are hole concentration, Boltzmann constant and electron charge, respectively. Using the p -values given in Table 1, the S values for the samples have been calculated and shown in Table 1. As it can be seen, all the values are negative, clearly indicating a dominating electron conduction mechanism.

In order to confirm it, thermoelectric power of the samples has been measured. Fig. 2 shows the temperature dependence of S for the K and Na substituted samples. All the samples exhibit both negative and positive S values which are similar to those observed for the different substituted BSCCO system by other groups [31-35,40]. The negative S values are observed from the room temperature down to what it is called the inversion temperature (below which S possesses positive values). The inversion temperatures of the pure, K01, K02, K03, Na01, Na02, Na03, Na04 and Na05 samples are found to be 183, 172, 201, 177, 205, 185, 197, 214 and 245 K, respectively. Below these temperatures S becomes positive. Furthermore, by decreasing the temperature, S increases and attain maximum values at around their respective transition temperature values [see Ref. [23,24] for T_c 's], before dropping to zero at lower temperatures, as expected. On the other hand, a change in the sign of S , as the temperature decreases, suggests the presence of both positive (holes) and negative (electrons) carriers and indicates the multiband nature of the compounds [40]. Positive contributions come from the holes and negative contributions from the electrons. The electron contribution to S may come from a strong energy dependence of the electron mobility, causing the negative S values at higher temperatures.

3.4. Thermal conductivity

The temperature dependence of thermal conductivity for all the samples is shown in Fig. 3. As can be observed in these figures, for $T > T_c$ (metallic state), κ linearly decreases with decreasing temperature, as expected. Around T_c , first a shallow minimum followed by a small raise and then almost a linear decrease towards zero was obtained with decreasing T . This type of behavior has previously been reported in the HTc systems [37,38,53,54]. The origin of the minimum can be attributed to the superconducting fluctuations [69,70]. On the other hand, it may be also due to an electronic contribution or a decrease on the phonon scattering mechanism or both [57,59] with increasing Na or K concentration.

These results reveal that the magnitude of the thermal conductivity, κ , is influenced by the K or Na-concentration in the system. Namely, with increasing Na concentration the magnitude of κ is decreased. But for the K substitution, the magnitude of κ is drastically increased for $x=0.75$.

3.5. Figure of Merit

The dimensionless figure of merit, ZT , is defined as:

$$ZT = \alpha^2 T / \rho \kappa \quad (4)$$

where T , α , ρ , κ are temperature, thermoelectric power, electrical resistivity and thermal conductivity, respectively. According to Eq. 4, ZT is maximized when the thermoelectric power increases and the thermal conductivity decreases. As can be seen from Fig. 4, ZT becomes zero at the so-called inversion temperature where thermoelectric power changes the sign, and it is maximized at about the material's transition temperature.

4. Conclusion

This study covers the XRD, carrier concentrations, thermoelectric power and thermal conductivity results of the $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{Na}_x\text{Cu}_2\text{O}_{8+y}$ superconductor with $x=0.05, 0.075, 0.10, 0.15,$ and 0.20 , and $\text{Bi}_2\text{Sr}_2\text{Ca}_{1-x}\text{K}_x\text{Cu}_2\text{O}_{8+y}$ superconductors with $x=0.05, 0.075, 0.10$ prepared by a polymer solution technique using polyethyleneimine, PEI, at temperatures between 20 K and 300 K. All the samples show both negative and positive thermoelectric power values as a function of temperature, changing their sign at the so-called inversion temperatures. The results indicate that, while at higher temperatures electrons are effective on the thermoelectric power, holes become effective at lower temperatures. The thermal conductivities of the samples show a shallow minimum followed by a sharp rise (or peak) just below their T_c values. In addition, the magnitudes of the thermal conductivities decrease with increasing Na

concentration but for the K substitution, they increase with increasing K-content ($x=0.75$). The rapid rise of κ , and hence development of a peak just below T_c , may be due to the enhancement of the quasi-particle contribution to the heat conductivity and to an increase of the quasi-particle mean free path in the superconducting state [57,58].

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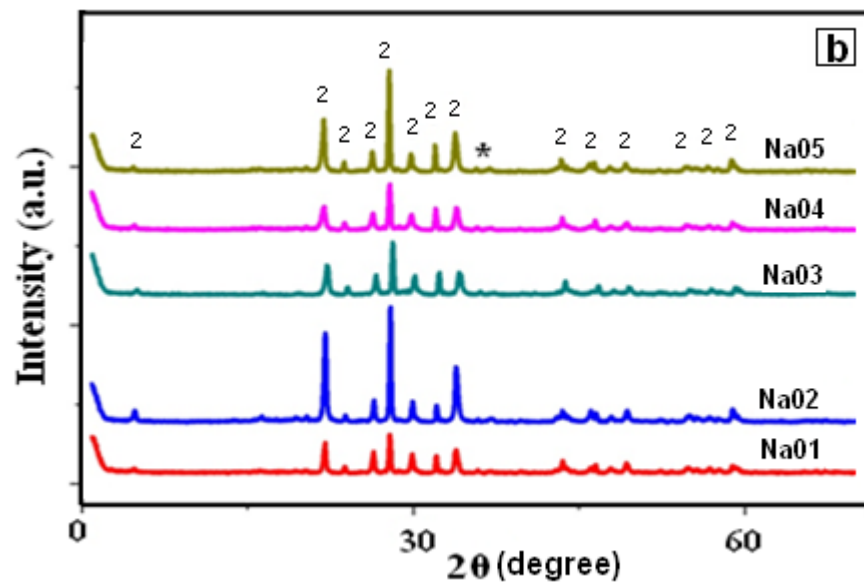
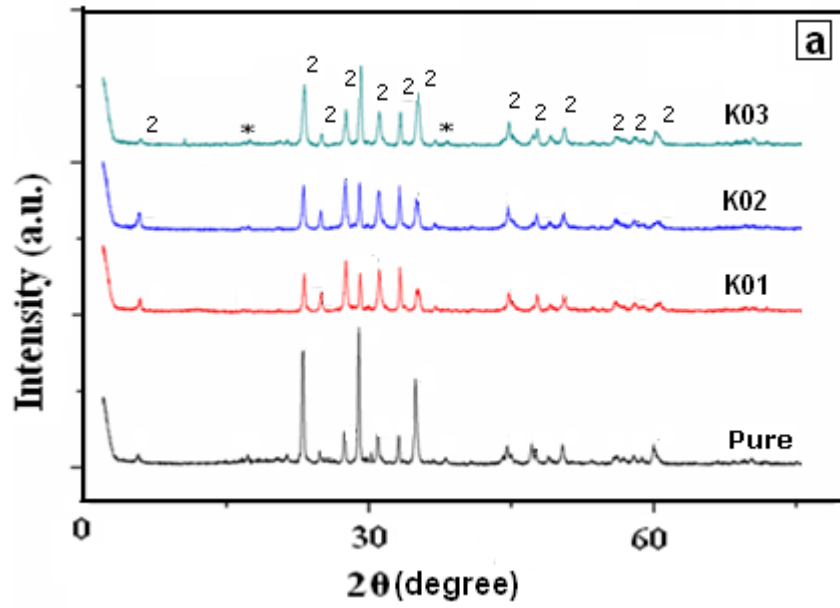


Fig.1. XRD patterns of Bi-2212 samples substituted with, a) K; and b) Na. Peaks corresponding to the Bi-2212 and CaCuO_2 phases are indicated by #2 and *, respectively

<i>Samples</i>	<i>Unit-cell parameter a=b (Å)</i>	<i>Unit-cell parameter c (Å)</i>	<i>L_{hkl} (Å)</i>	<i>Hole Concent. (p)</i>	<i>S(10⁻⁴ V/K)</i>
<i>Pure</i>	5.4136	30.8657	375.64	0.1333	-1.775
<i>K01</i>	5.4153	30.8542	385.96	0.1336	-1.784
<i>K02</i>	5.4161	30.8276	398.42	0.1406	-2.033
<i>K03</i>	5.4183	30.7306	394.14	0.1257	-1.558
<i>Na01</i>	5.4283	30.7530	432.68	0.1811	-7.026
<i>Na02</i>	5.4336	30.7330	445.44	0.1786	-6.173
<i>Na03</i>	5.4391	30.7322	446.85	0.1807	-6.874
<i>Na04</i>	5.4415	30.7318	450.45	0.1787	-6.204
<i>Na05</i>	5.4494	30.7290	463.68	0.1730	-6.834

Table-1: Unit cell parameters, crystal size, hole concentration and calculated thermoelectric power values for each sample

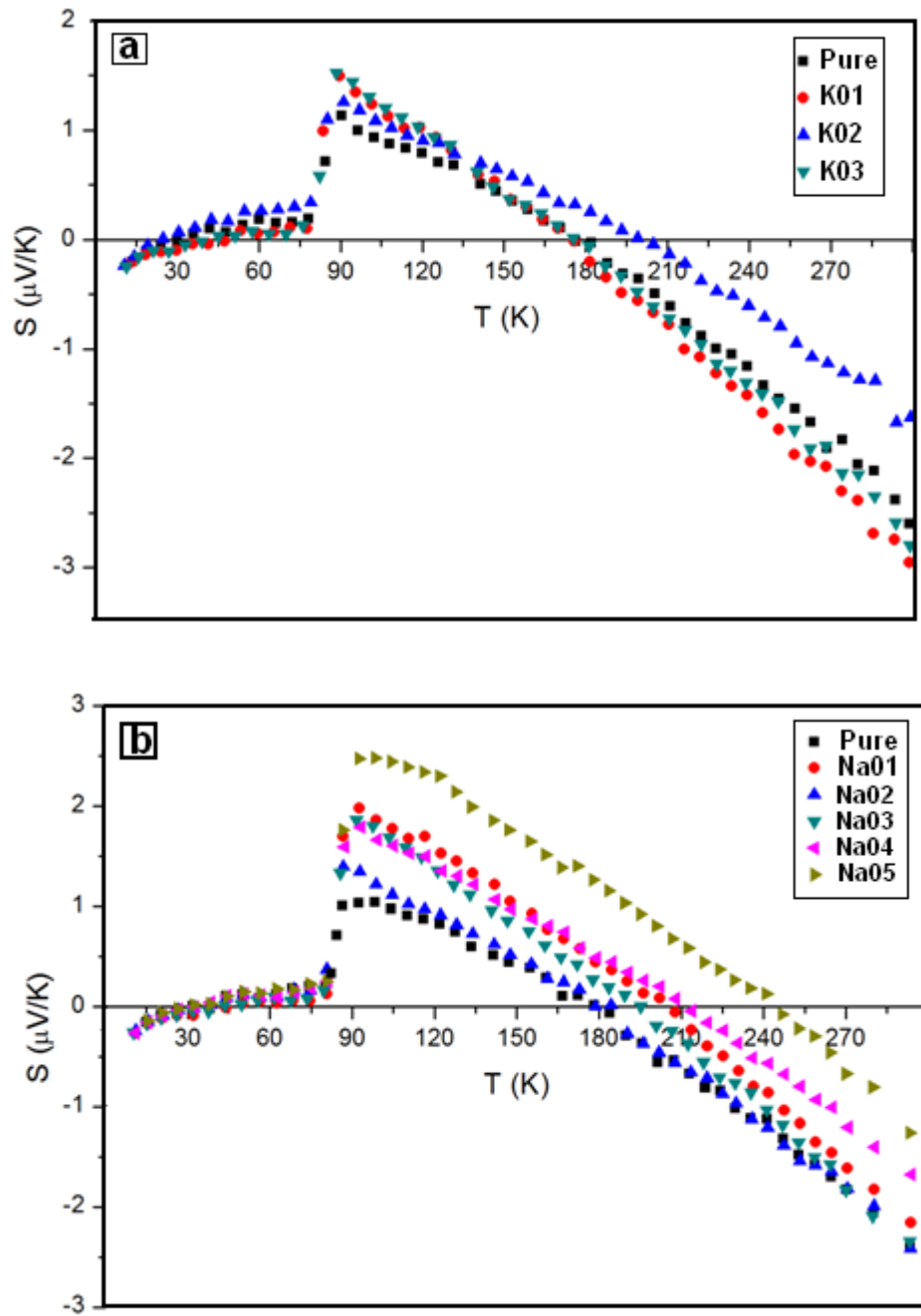


Fig.2: Temperature dependence of thermoelectric power for the samples substituted with K (a); and Na (b).

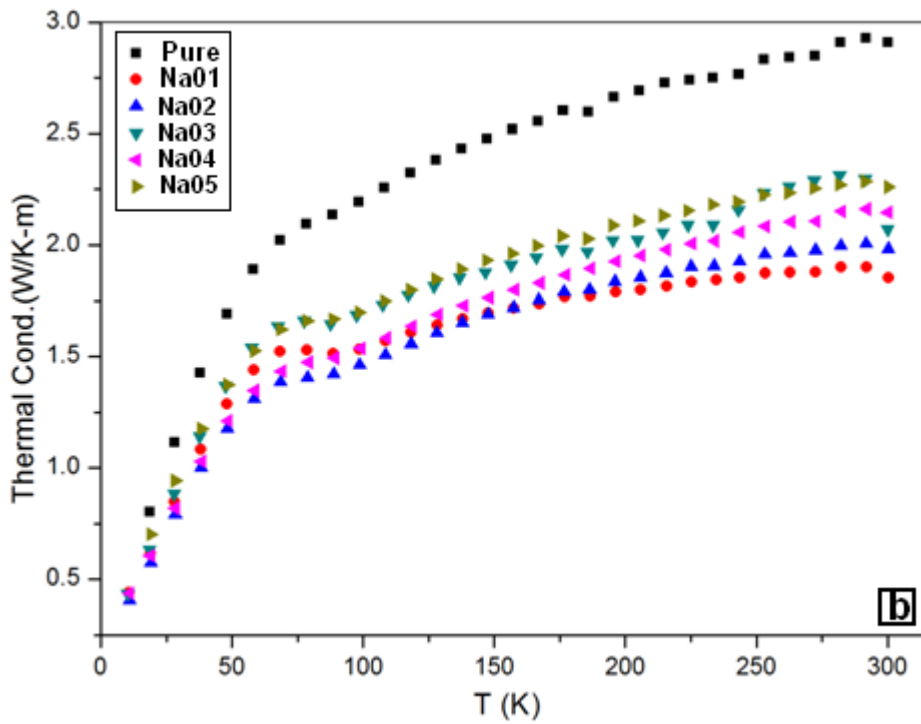
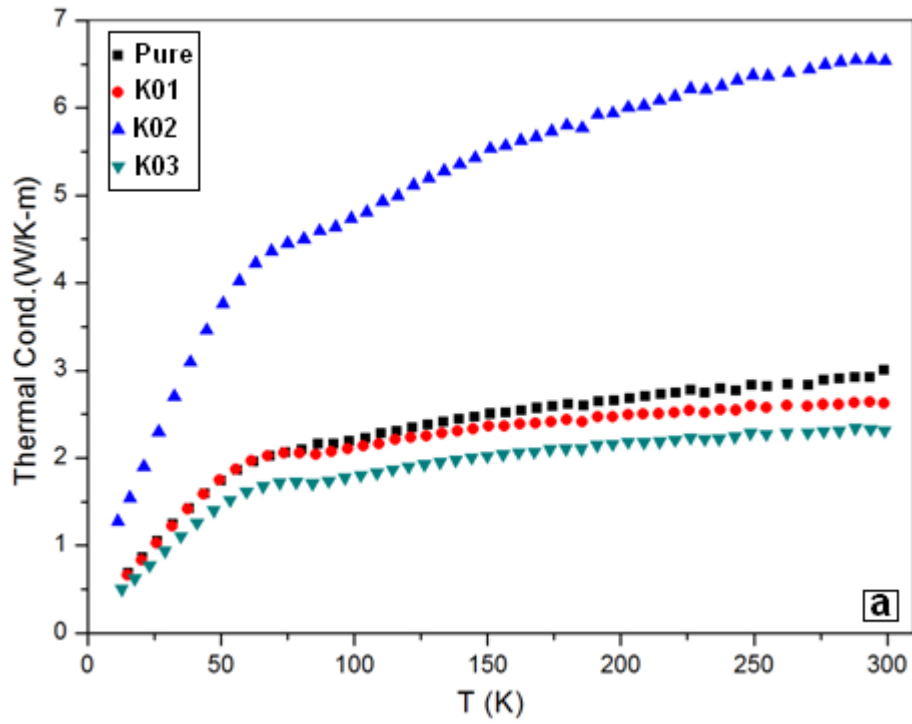


Fig.3: Temperature dependence of the thermal conductivity for the K-doped (a); and Na-doped (b) samples.

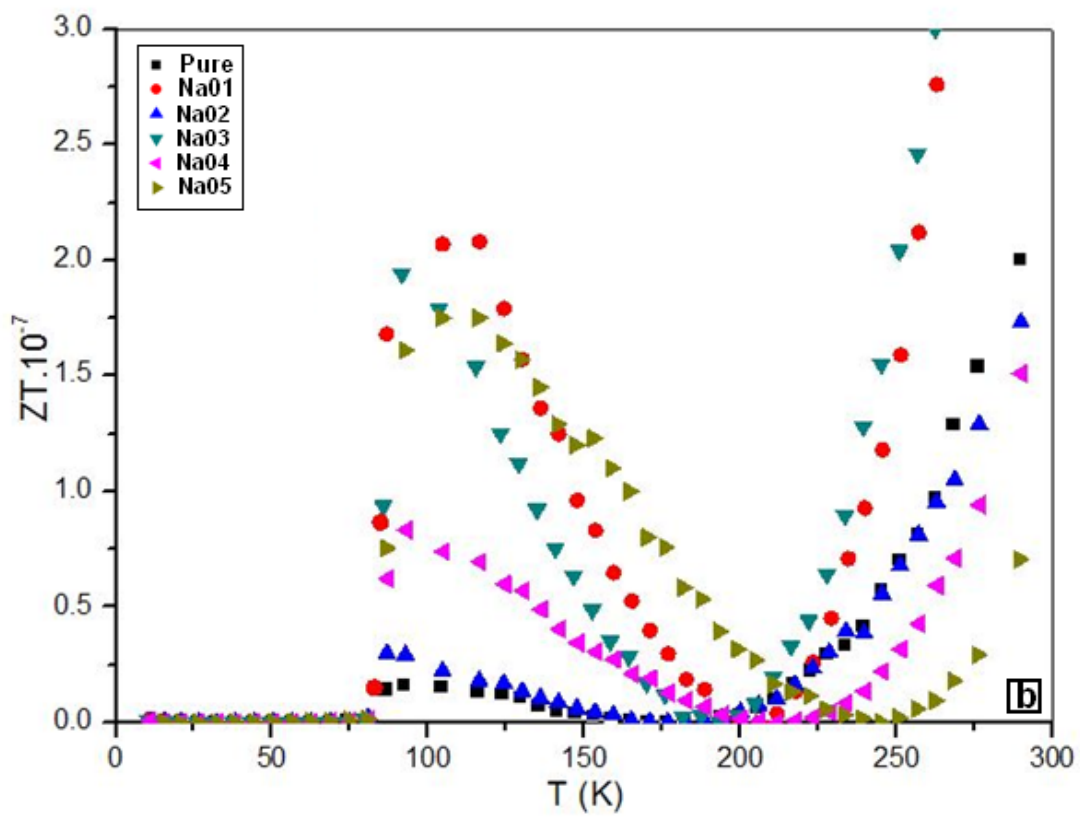
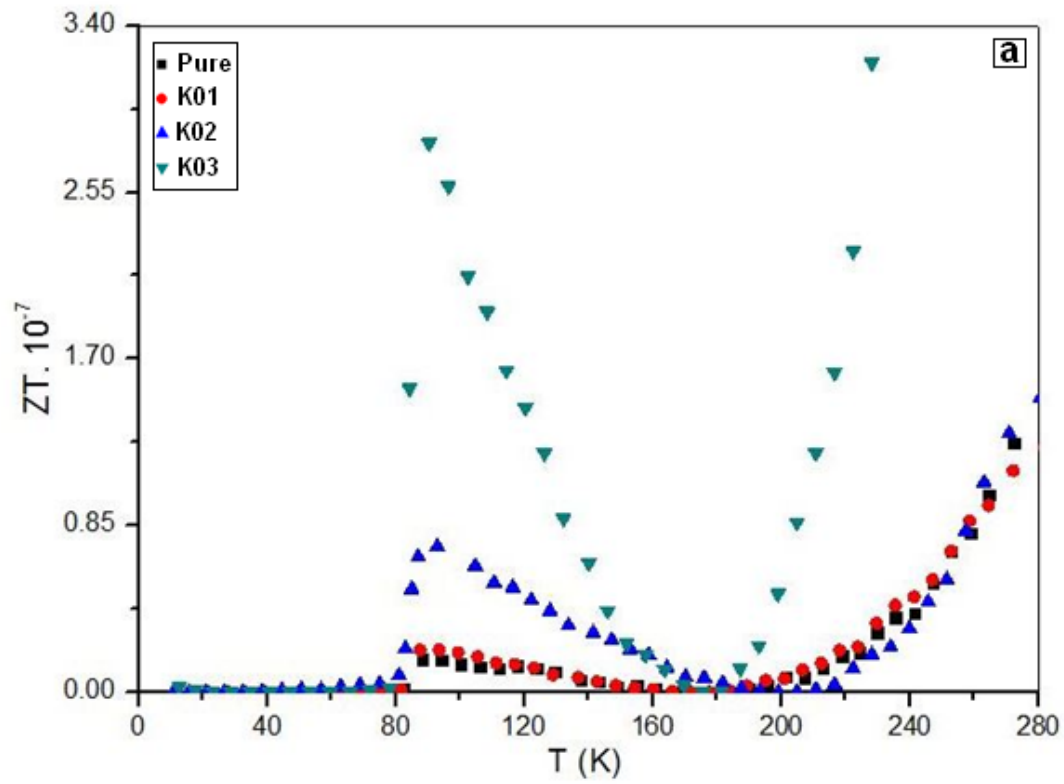


Fig.4: Figure of Merit for the K (a); and Na (b) Bi-2212 substituted samples.