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Comparison of the superconducting properties of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ prepared at different synthesis temperatures

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Abstract

The superconducting properties of Zn-doped $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ {CuTlZn-1223} ($y=0$, 0.83, 1.66, 2.5) samples prepared at 820, 830, 850 and 860 °C have been compared. The samples were investigated by x-ray diffraction (XRD), dc-resistivity, ac-susceptibility and Fourier Transform Infrared (FTIR) absorption measurements. Almost all the superconducting properties have been increased to their maximum in all CuTlZn-1223 samples synthesized at 860 °C, which shows that 860 °C is the optimum temperature to achieve CuTlZn-1223 with enhanced superconducting properties.

Keywords: $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_3O_{10-\delta}$ superconductors; Synthesis temperature; Zn-doping

1. Introduction

In oxide high T_c cuprates, the substitution of impurity atoms has been used as a probe to understand the mechanism of high temperature superconductivity [1,2]. The substitution of non-magnetic Zn^{+2} at Cu^{+2} sites in CuO_2 planes has been found to suppress the superconductivity [3–31]. It is common consensus that in oxide high T_c superconductors, the superconductivity phenomenon takes place in CuO_2 planes and the free carriers interact with the outer most $3d^9$ electrons of the copper atoms, which ultimately impart d-wave symmetry to the carriers in their respective conduction bands [32]. Most of these oxide high T_c superconductors are an-isotropic and their conductivity parallel to CuO_2 planes (i.e. along '*a*' and '*b*' axes) is much higher than that of normal to these planes (i.e. along *c*-axis). The an-isotropy ($\gamma=\xi_c/\xi_{ab}$) of these compounds increases with the increase in the number of CuO_2 planes. The addition of CuO_2 plane beyond $n=2$ (n is number of CuO_2 plane) increases the magnitude of anti-ferromagnetism in these oxide superconductors because the additional inner CuO_2 planes are in the under-doped region. The doping of Zn^{+2} at Cu^{+2} sites in CuO_2 planes in d-wave high temperature superconductors (HTSC's) was suggested to be the source of pair-breaking mechanism and electronic localization in the neighborhood of Zn-doped atoms [33–50]. It was suggested that in $YBa_2Cu_3O_{7-\delta}$ superconductor, localization of carriers at Cu^{+2} sites in the CuO_2 planes in the immediate vicinity of Zn^{+2} suppresses superconductivity [12].

Contrary to all the previous studies on Zn-doped cuprate HTSC's, we have observed enhanced superconductivity after Zn-doping in $Cu_{0.5}Tl_{0.5}Ba_2Ca_{n-1}(Cu_{n-y}Zn_y)O_{2n+4-\delta}$ ($n=3$, 4; $y=0$, 0.5, 1.0, 1.5, 2.0, 2.5, for $n=3$ and $y=0$, 0.5, 1.0, 1.5, 2.0, 2.5, 3.0, 3.5 for $n=4$) superconductors [51,52]. We did not observe the localization of the carriers at the Cu^{+2} and Zn^{+2} sites in these compounds by carrying out the post-annealing experiments in air, nitrogen and oxygen.

Since the choice of synthesis temperature is also a critical parameter to achieve the material with superior superconducting properties, therefore, in the present work we have synthesized $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ superconductor samples at 820, 830, 850 and 860 °C temperatures and

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compared their superconducting properties. We have presented the results of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples synthesized at 820 and 830 °C in this article. These studies would help to find the best synthesis temperature for $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ superconducting phase with higher superconductivity.

2. Experimental

The ceramic superconducting $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) material was prepared by the solid-state reaction method accomplished in two stages. At the first stage $Cu_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ precursor material was prepared by thoroughly grinding Cu(CN), $Ba(NO_3)_2$, $Ca(NO_3)_2$ and ZnO in a quartz mortar and pestle in appropriate ratios. The mixed material was fired twice at 820 °C in a quartz boat for 24 h and furnace was cooled to room temperature. At the second stage, the precursor material was mixed with Tl_2O_3 and ground for about an hour. Thallium oxide mixed material was pelletized under 3.8 t/cm^2 and the pellets were enclosed in a gold capsule. The pellets in the gold capsule were sintered at 820 °C for about 10 min and then quenched to room temperature to get $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ composition.

These samples were characterized by dc-resistivity, ac-susceptibility and Fourier Transform Infrared (FTIR) absorption measurements. The structure of the material was determined by x-ray diffraction (XRD) scan from Rigaku D/Max IIIC, using a CuKα source of wavelength 1.54056 Å. The cell parameters were determined by a cell refinement computer program. The phonon modes related to the vibrations of various oxygen atoms in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples were observed by Nicolet 5700 Fourier Transform Infrared Spectrometer (FTIR) in the wavenumber range of 400–650 cm^{-1}. The $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples were also synthesized at 830, 850 and 860 °C following the similar procedure [51].

3. Results and discussion

3.1. $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples synthesized at 820 °C

The x-ray diffraction (XRD) patterns of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) samples have shown tetragonal structure, Fig. 1. After fitting the x-ray diffraction lines to the tetragonal structure for $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples, the cell parameters (*a* and *c*-axes) calculated are 3.92, 4.05, 4.48, 4.81 and 15.4, 15.1, 14.88, 14.84 Å for y=0, 0.83, 1.66, 2.5 respectively. The volumes of the unit cells corresponding to these axes lengths are 236.6, 247.7, 298.6, 343.3 Å^3 respectively.

In $Cu_{0.5}Tl_{0.5}Ba_2Ca_{n-1}(Cu_{n-y}Zn_y)O_{2n+4-\delta}$ (n=3, 4) high temperature superconductor samples synthesized at 860 °C (for n=3) and 880 °C (for n=4), the apical oxygen phonon modes of type Tl–O_A–M(2) and Cu(1)–O_A–Cu(2) were observed in the wave number range 454–458 and 514–514 cm^{-1} and the CuO_2 planar oxygen mode around 565–575 cm^{-1} [51–52]. The FTIR absorption measurements of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples are shown in Fig. 2. The apical oxygen modes of type Tl–O_A–M(2) and Cu(1)–O_A–Cu(2) have been shifted to lower wave number after Zn-doping from 460, 520 cm^{-1} to 454–457, 517–518 cm^{-1} respectively. The CuO_2 planar oxygen modes in Zn free samples which were observed around 549–567 cm^{-1}, have been systematically softened to 548–565 cm^{-1} with the increased incorporation of Zn (i.e y=0.83, 1.66, 2.5).

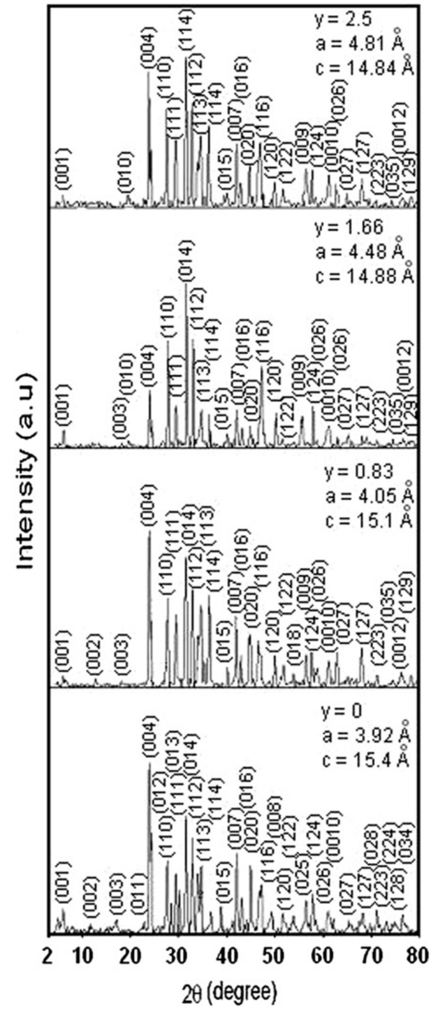

Fig. 1. X-ray diffraction (XRD) patterns of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) superconductor samples synthesized at 820 °C temperature.

The resistivity versus temperature measurements of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ superconductor samples are shown in Fig. 3. All the samples have shown metallic variations of resistivity from room temperature down to onset of superconductivity. These samples have shown onset of superconductivity around 118, 111, 114, 112 K and zero resistivity critical temperatures $T_c(R=0)$ around 101, 98, 92, 93 K for y=0, 0.83, 1.66, 2.5 in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples. The ac-susceptibility measurements of these samples are shown in Fig. 4. The onset temperatures of

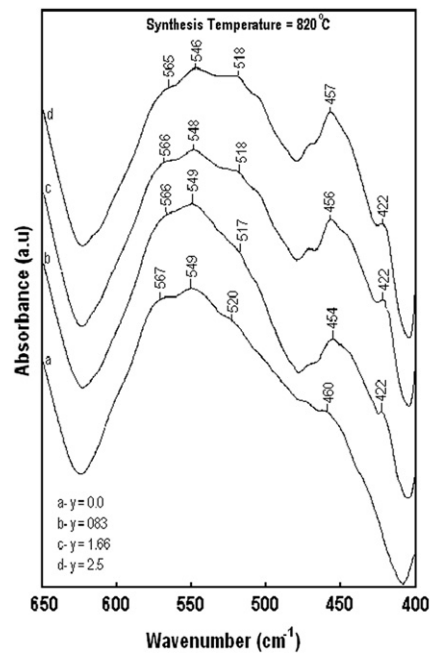

Fig. 2. FTIR absorption measurements of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) superconductor samples synthesized at 820 °C temperature.

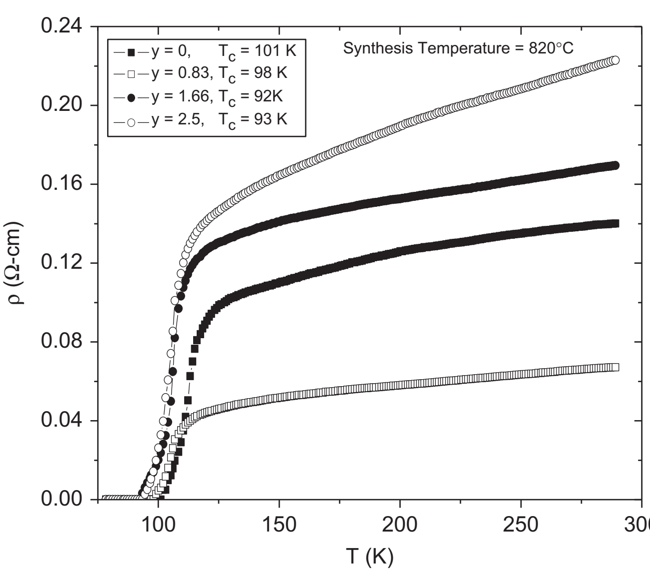

Fig. 3. The resistivity vs temperature of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) superconductor samples synthesized at 820 °C temperature.

diamagnetism are observed around 102, 100, 98, 97 K and the peak temperatures (T_P) in the out-of-phase component of magnetic ac-susceptibility around 97, 96, 94, 93 K, respectively.

3.2. $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples synthesized at 830 °C

The x-ray diffraction patterns of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) samples synthesized at

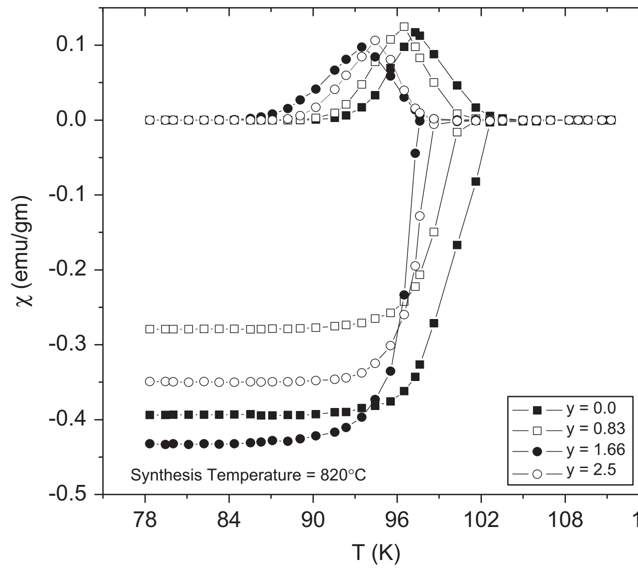

Fig. 4. The ac-susceptibility vs temperature of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) superconductors samples synthesized at 820 °C temperature.

830 °C have shown tetragonal structure, Fig. 5 (a and b). The cell parameters (a and c-axes) calculated after fitting the data to the tetragonal structure for $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ samples for a and c-axes are 3.92, 4.15, 4.22, 4.37 Å and 15.03, 14.99, 14.84, 14.82 Å respectively. The volume of unit cell corresponding to these axes lengths are 230.2, 258.16, 264.27, 283.01 Å^3 respectively. The volume of the unit cell of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ samples synthesized 830 °C temperature is lower than that of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ samples synthesized 820 °C temperature. The possibility of lower Tl-content in 830 °C prepared sample is most likely the source of decreased volume of the unit cell.

The FTIR absorption measurements of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) samples synthesized at 830 °C are shown in Fig. 6. The apical oxygen band of the type Tl–O_A–Cu(2) in Zn free samples were observed around 428–487 cm^{-1}. The relative intensity of that mode has been increased and softened to 422~480, 425~463, 423~465 cm^{-1} in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ for y=0.83, 1.66, 2.5 samples respectively. The vibrations of apical oxygen mode of type Cu(1)–O_A–Cu(2) in Zn free samples which were observed in the range of 528–550 cm^{-1}, have been shifted to lower values of wavenumber 509–545, 514–550, 511–545 cm^{-1} in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ samples for y=0.83, 1.66, 2.5 respectively. The CuO_2 planar modes in Zn free samples observed around 573 cm^{-1}, have been moved towards lower wavenumber around 572, 568, 564 cm^{-1} in Zn-doped $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ samples. The softening of all oxygen modes is most likely linked with the increased mass of Zn (65.38 amu) as compared to that of Cu (63.54 amu) and suppression of Jahn-Teller distortions with the increased Zn-doping in the final compound [52]. The Cu^{+2} ions show a strong Jahn-Teller effect; the octahedron coordination of Cu^{+2} in the planes

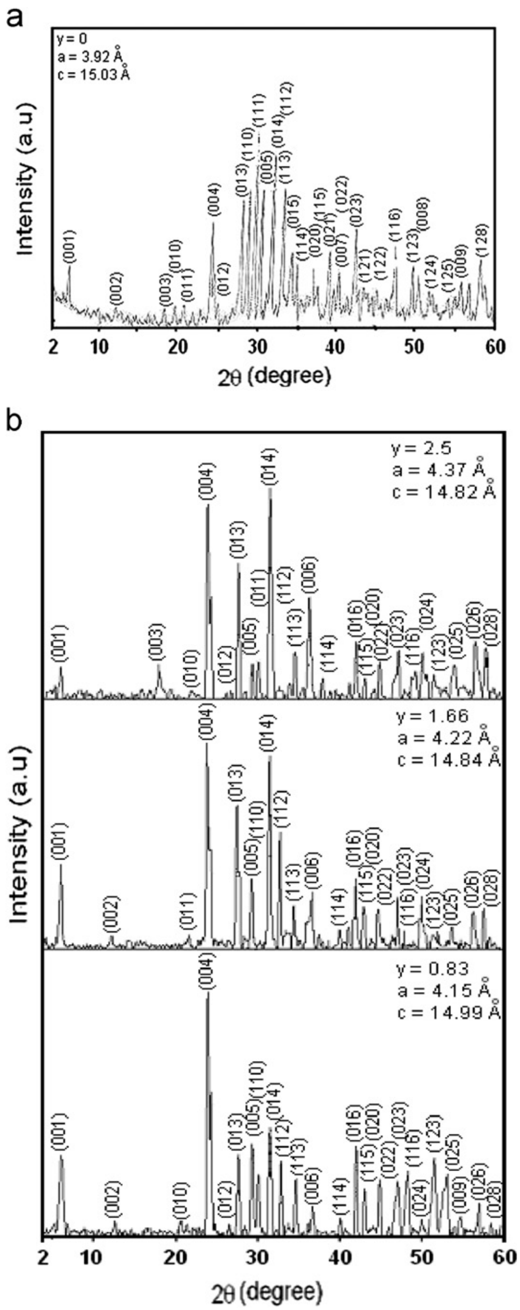

Fig. 5. (a) X-ray diffraction (XRD) pattern of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_3O_{10-\delta}$ superconductor sample synthesized at 830 °C temperature. (b) The x-ray diffraction (XRD) patterns of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ (y=0.83, 1.66, 2.5) superconductor samples synthesized at 830 °C temperature.

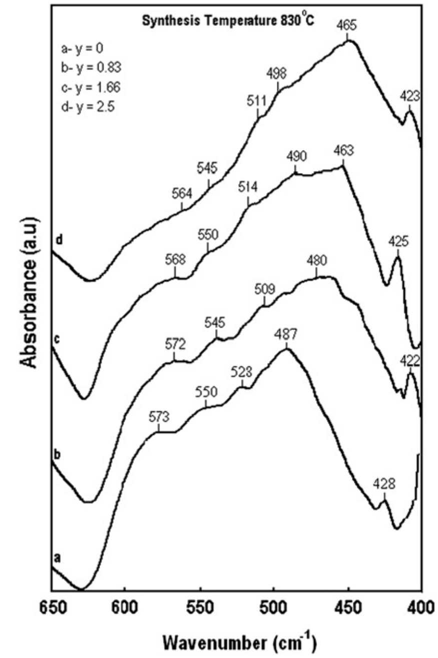

Fig. 6. The FTIR absorption measurements of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) superconductor samples.

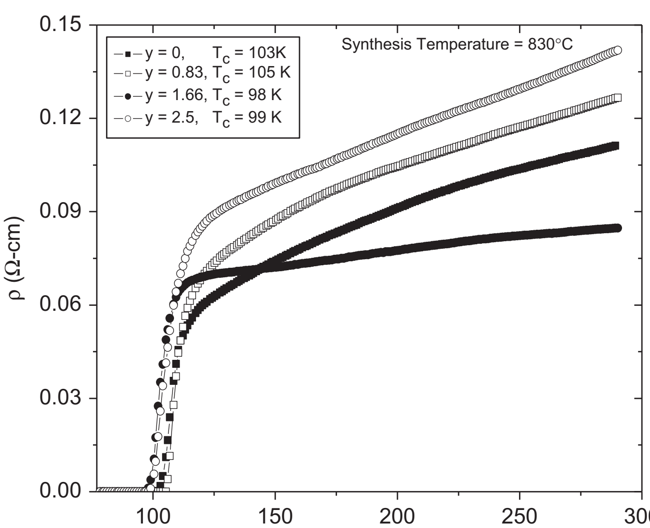

Fig. 7. The resistivity vs temperature of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) superconductor samples synthesized at 830 °C temperature.

are elongated along *c*-axis [26]. However, the octahedrons around Zn^{+2} do not exhibit distortions around Zn^{+2} in the plane because they are in the $3d^{10}$ state; the lowest energy spin-less state. Hence, the doping of Zn^{+2} at Cu^{+2} sites reduces the local Jahn-Teller distortions.

The Jahn-Teller distortion is interaction of spin lattice with the Fermi Sea of mobile carriers. In other words afore-mentioned distortions are suppressed and the decreased spin–spin scattering would promote a decrease in the *c*-axis length and it was expected that decreased *c*-axis length would promote hardening of apical oxygen modes; contrary to this we have observed their softening in samples that was only due to heavier Zn^{+2} (65.38 amu) as compared to that of Cu^{+2} (63.54 amu).

The resistivity measurements of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ (y=0, 0.83, 1.66, 2.5) samples synthesized at 830 °C are shown in Fig. 7. These samples have shown metallic variations of resistivity from room temperature down to onset of superconductivity. The onset temperatures of

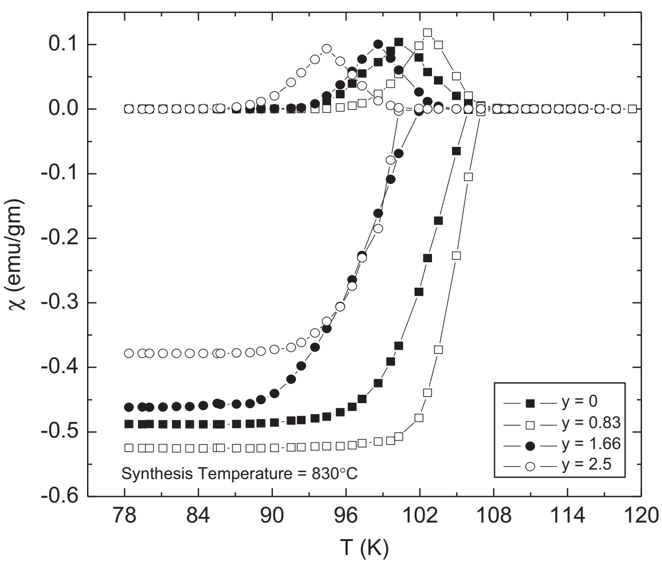

Fig. 8. The ac-susceptibility vs temperature of $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ ($y=0$, 0.83, 1.66, 2.5) superconductors samples synthesized at 830 °C temperature.

superconductivity in these samples are observed around 112, 116, 109, 113 K and zero resistivity critical temperature T_c ($R=0$) around 103, 105, 98, 99 K for $y=0$, 0.83, 1.66, 2.5 in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ samples. The magnetic ac-susceptibility measurements of these samples are shown in Fig. 8. The onset temperatures of diamagnetism observed in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2Cu_{3-y}Zn_yO_{10-\delta}$ samples are around 105, 106, 102, 100 K and the peak temperatures (T_P) in the out-of-phase component magnetic ac-susceptibility around 100, 102, 98, 94 K, respectively.

4. Conclusion

The effects of synthesis temperature on superconductivity in Zn-doped $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ ($y=0$, 0.83, 1.66, 2.5) samples have been investigated to select the best synthesis temperature for $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ superconducting phase. The synthesis of Zn-doped $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ superconductor samples at 820 and 830 °C temperatures in present studies and at 850 and 860 °C in the previous findings have shown that $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ superconducting phase can be synthesized over a wide temperature window with different Tl-content in $Cu_{0.5}Tl_{0.5}Ba_2O_{4-\delta}$ charge reservoir layer. The suppression of superconductivity in $Cu_{0.5}Tl_{0.5}Ba_2Ca_2(Cu_{3-y}Zn_y)O_{10-\delta}$ samples synthesized at relatively lower temperature is most likely due to higher Tl-content in $Cu_{0.5}Tl_{0.5}Ba_2O_{4-\delta}$ charge reservoir layer. At higher synthesis temperature beyond 860 °C, the higher order CuTl-1234 superconducting phase formation takes place. So, it is concluded that the best synthesis temperature for $Cu_{0.5}Tl_{0.5}Ba_2Ca_2$ $(Cu_{3-y}Zn_y)O_{10-\delta}$ superconducting phase is 860 °C at which the optimum superconducting properties have observed.

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