Synthesis and Specification of Superconductor Compound $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4+\delta}$(n = 2, 3, 4)

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ABSTRACT

High-temperature superconductor compound $Tl_2Ba_2Ca_{n-1}CunO_{2n+4+\delta}$ *(n* = 2, 3, 4*)* was prepared by solid-state reaction evacuated sealed tube method. The research was carried out to investigate the effect of parameter (*n*) on the superconductor behavior of the compound $Tl_2Ba_2Ca_{n-1}CunO_{2n+4+\delta}$. The structural phase of prepared samples was analyzed by X-ray diffraction (XRD) analysis, and the results showed the demand phase depend on the parameter (*n*). The resistivity measurement by four-probe technique used to investigate the critical temperature. The parameter (*n*) has major effect on definition of critical temperature for superconductor compound. The results showed that the sample prepared with *n* = 2 has a critical temperature T_c= 102 K; meanwhile, the sample prepared with *n* = 4 has a critical temperature T_c= 99 K; however, the sample which was prepared with *n* = 3 has a maximum critical temperature T_c= 121 K. There is a match between the result of magnetic susceptibility and the resistivity measurements at *n* = 3.

Keywords X-ray diffraction, Resistivity measurement ,Tl-family superconductor, Lattice constants.

تحضير وتصنيف المركب الفائق التوصيل $Tl_2Ba_2Ca_{n-1}CunO_{2n+4+\delta}$*(n* = 2, 3, 4*)*

الخلاصة

يتضمن البحث تحضير ودراسة مادة فائقة التوصيل (Tl2Ba2Can−1CunO2n+4+δ(n = 2, 3, 4) على شكل مركب سيراميكي بطريقة تفاعل الحالة الصلبة بوجود الاوكسجين وفي الكبسولة (انبوب كوارتز مفرغ من الهواء). وتم أخذ قيم 2 ، 3 ، 4 للمعامل (n) . وتمت دراسة هذه العينات بواسطة تأثير ميزنر وقياس محتوى الاوكسجين المضاف خلال عملية التلبيد عن طريق الفحص الكيميائي(المعايرة باستخدام اليود) وتحليل بواسطة حيود الاشعة السينية لدراسة التركيب البلوري للمركب وايجاد الدرجة الحرجة للمركب وتحليل الطيفي لطاقة التشتت للعناصر بواسطة الاشعة السينية وقياس المقاومة الكهربائية بتقنية المتحسسات الاربعة لقياس درجة الحرارة الحرجة للمركب والقابلية المغناطيسية للمركب. ولقد اظهرت النتائج ان المركب (n=2) قيمه درجة الحرارة الحرجة كانت بحدود 102(K) وان المركب (n=3 ،) قيمة درجة الحرارة الحرجة بحدود)121 (K بينما درجة الحرارة الحرجة للمركب (n=4 ,) كانت بحدود 99 (K) . أوضحت نتائج تحليلات حيود ألاشعة السينية (XRD) للقيم (n=2,3,4) للمركب السيراميكي لها تركيب

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رباعي الاطوار(Tetragonal) وقيمة البعد البلوري c-axis تزداد بزيادة عدد الطبقات للعناصر $CaCuO_2$.

ولقد أظهرت نتائج القابلية المغناطيسية أن درجة الحرارة الحرجة للمركب السيراميكي تطابق تام مع نتائج وقياس المقاومة الكهربائية بتقنية المتحسسات الاربعة لقياس درجة الحرارة الحرجة للمركب.

INTRODUCTION

Superconductivity in high-Tc superconductors is generally regarded as a property of CuO_2 layers. The number n of CuO_2 layers per chemical formula unit may vary within a homologous series: For $Tl_2Ba_2Ca_{n-1}CunO_{2n+4+\delta}$ bulk samples with n=1, 2, 3, and 4 have been synthesized. In then=2 compound both CuO2 layers per half unit cell are crystallographically equivalent However, in the n=3 and n=4compounds, there are two different kinds of CuO2 layers: the inner (i) CuO2 layers and the outer (o) CuO2 layers which are crystallographically in equivalent.[1-3] In the Tl-based high temperature superconductor (HTS) family, $Tl_2Ba_2Ca_2Cu_3O_{10}$ (Tl-2223) are attractive due to their high critical temperatures of up to 120 K.[4] The critical temperature suggests that the Tl-2223 could be operated at higher temperature. As a result, electronic devices can be fabricated in a compact planar form, offering a significant saving in size and weight, which makes it possible to operate at temperatures over 100 K and to reduce the cost of refrigeration devices for microwave systems.[5]The final product of the superconducting samples and their properties are directly influenced by method used [6].In this research, we have prepared different samples like (Tl-2212), (Tl-2223), and (Tl-2234) phases using solid-state reaction with a certain condition of calcinations and sintering depends on *n*. The samples have been characterized by X-ray diffraction (XRD) and measurement of resistivity versus temperature and Magnetic susceptibility.

Experimental Procedure

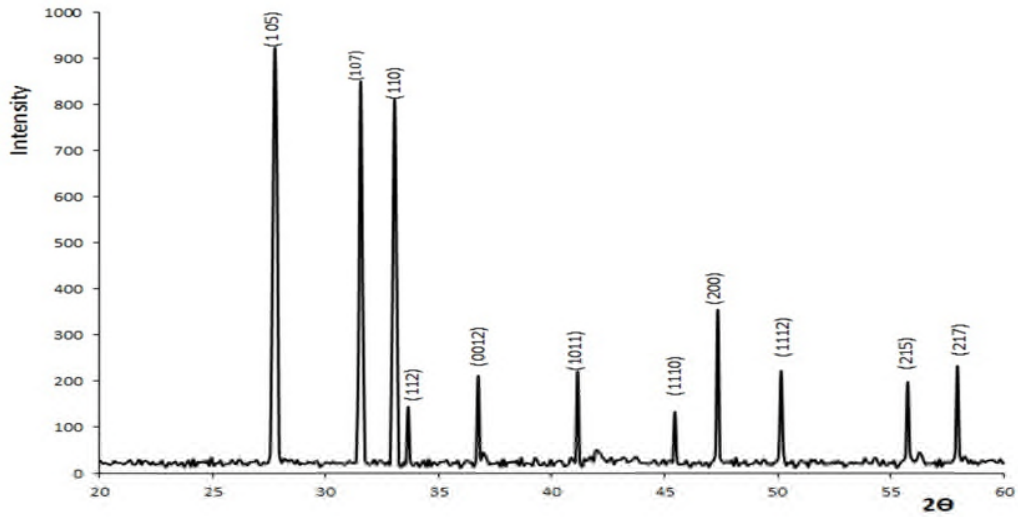
High-temperature superconductor system Tl–Ba–Ca–Cu–O was prepared by using solid-state reaction method with appropriate quantities of high purity of Tl_2O_3, BaO, CaO, and CuO. The mixture was grinding manually with 2- propanol using hand mortar. The well-mixed powder of these oxides and carbonates was calcined at 840 ◦C for 50 h with five intermediate grinding for each 10 h, The calcined powder were regrind again and pressed into pellet shaped with 1 cm in diameter and 0.1 cm thick The benefit of multicalcinations process is to create the superconducting phase and remove undesired elements from the mixture, whereas the sintering treatment tend to reduce the pour size and make the samples denser in order to enhance the superconducting behavior. Sample with n = 2 was sintered at 840 ◦C in oxygen flow with rate 20 % for 20 h; the sample with n = 3 was sintered at 860 ◦C in oxygen flow with rate 20 % for 7 h; and sample with n = 4 was sintered at 850 ◦C in oxygen flow with rate 5 % at 15 h The heating rate was 300◦C/h whereas the cooling rate to room temperature was 300◦C/h. The sintering process was in evacuated sealed tube The X-ray diffraction with $CuK\alpha 1$ radiation (λ = 1.5406 A°) was used to show the presence of superconducting phase. The resistivity and magnetic susceptibility was a good tool to investigate the T_c value and which value of n is preferable in the superconducting properties.

Results and Discussion

After the successive preparation of superconductor samples that was investigated by the Meissner effect, later, the major analysis and measurements have been done such as X-ray diffraction, resistivity, and magnetic susceptibility measurements. The analysis of X-ray diffraction pattern for $Tl_2Ba_2CaCu_2O_{8+\delta}$ (n=2) by using software called Match and JCPDS (International Center for Diffraction Data) and the number of the PDF file [80-406] are shown in Fig. 1. It was found that the structural phase was tetragonal phase. The concluded lattice constants were found *a* = b= 3.842 A° and c= 29.39 A° , and the volume of the unit cell was about 435.55 $A^{\circ 3}$ as shown in Table 1. There is a slightly variation of the lattice constant calculated in comparable with the standard values [80-406]. That is, return to a slightly shift in such diffracted peaks (2θ = 27.75, 31.55, 33.05) with the miller indices (105), (107), and (110) toward the Higher angles were observed The diffraction pattern for $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$(*n* = 3), as shown in Fig. 2, the matching analysis with ICDD and ASTM card [80-0779] exhibited the structural phase was Tetragonal with lattice constant *a* = b =3.839 °A, and *c* = 35.78 °A and the volume of the unit cell was nearly 530.11 $A^{\circ 3}$. The highest maximum three peaks at 2θ = 29.15, 32.45, and 33.05. Normally, there is a shift in peaks toward the higher angles. This is the reason why there is a limited change in the lattice constant with standard values obtained by in the PDF file [80-0779]. The Miller indices for these angles were (107), (109), and (110) respectively. X-ray diffraction pattern for $Tl_2Ba_2Ca_3Cu_4O_{12+\delta}$(*n* = 4), as shown in Fig.3 had a tetragonal phase with the lattice constant *a* = *b* = 3.837 A° and *c* = 41.2 A° , and the volume of the unit cell was about 615.66 $A^{\circ 3}$. The shifting in peaks gives more details about the slightly change in the lattice constant with standard one [80-0780]. The highest three maximum peaks at 2θ = 30.35, 32.95 and 33.35 with Miller indices were (109), (110), and (112) respectively. X-ray diffraction peaks with Miller indices (107), (110), (112), and (200) appeared in all different compositions (n= 2,3,4). We can expect that all these peaks refer to a plane that contains Tl and Ba ions in the structure, because both are common for all concentrations with the values *n* = 2,3,4; other peaks refer to CaCuO planes in the unit cell with different value of (n). The prepared samples of Tl-2212, Tl-2223, and Tl-2234 all peaks were shifting to a higher angle side, which was the principle factor for the change in the lattice parameter. It might be affected by the O_2 concentration that was changed from the theoretical value toward the non-stoichiometric. The summarized data of XRD analysis is explained in Table 1,it was clear that the value *n* = 4 has the highest volume with Tetragonal system. There is a relationship between the lattice constant and the number of $CaCuO_2$ layers inserted as a function of increasing *n*. The results showed when *n* = 2 has a tetragonal phase; later, when increasing the number of $CaCuO_2$ layers at *n* = 3, there is a linear increase in the *c*-axis and still continue when *n* = 4. That means there is an elongation in the *c*-axis direction with constant values of the *a* and *b* axes. This result showed that there is an increase in the $CaCuO_2$ layers as a function of *n*. On the other hand, the parameters *a* and *b* started from a fixed value for *n* = 2,3 going to abrupt decrease at *n* = 4. This might be attributed to the inversion in the structure due to the shrinkage in the basal plane. The interpretation for that is the reduction of the vacancies or oxygen sites in the basal plane, which was confirmed with concluded low T_c. On the other hand, the ratio Ca/Cu is varied and proportional to the *n* value. That is insured that the variation of Cu ions takes more substituted site with respect to Ca ions in the structure. The O ions were remained at constant value that might be attributed to constant flow ratio for the oxygen inserted in the samples under study. The resistivity measurement of $Tl_2Ba_2CaCu_2O_{8+\delta}$ (n = 2), as shown in Fig. 4, was clear the region in resistivity curve; the exhibited of T_c with a certain value T_c= 102K. This is a proof that there is a phase in producing $Tl_2Ba_2CaCu_2O_{8+\delta}$because it contains high-T_c phase, whereas the result of resistivity curve for n = 3 as shown in Fig. 5. The results exhibited that the value of critical temperature was about121 K. On other hand, the effect of (n = 4) tend to reduce T_c to a value (99 K), as shown in Fig. 6. For both n = 3,4,this is a proof that there is a single phase because only one stage in the resistivity curve appeared. That is confirmed with XRD results. The lattice constants for the composition Tl-2234 was different in comparison with Tl-2223 and Tl-2212 as we mentioned before. The tetragonal unit cell was so small that it attributed to a small grain size which tends to produce a small pour size. This is the reason why the normal resistivity has a minimum value of about 2 Ωmm in comparison for both the compositions Tl-2212 and Tl-2223; they have 3 and 2.5Ωmm respectively. The maximum value of the critical temperature for Tl–Ba–Ca–Cu–O system, when n = 3 has a critical value of 121 K, may be attributed to decrease the structural defects and also due to the intergrowth of large number of Cu–O layer within the unit cell. The increase in T_c value as a function of n might be attributed to the increase in the density of charge carriers, which are responsible on the conductivity mechanism. The result of magnetic susceptibility of $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$was measured at different temperature range 50–185 K as shown in Fig. 7. The curve showed that diamagnetic transition temperature was about 122 K. This T_c value is compatible with the value obtained from Fig. 5. That means there is a good agreement between the results of XRD, resistivity, and Magnetic susceptibility measurements. The magnetic susceptibility was about −0.5, and the negative values means the magnetization in opposite direction of the magnetic field.

Table (1) Variation of lattice constants (a,b,c), volume, and structural phase with different values of n

Formula	n value	a(A^0)	b(A^0)	c(A^0)	V(A^{03})	Structural phase
$Tl_2Ba_2CaCu_2O_{8+\delta}$	2	3.842	3.842	29.39	435.55	Tetragonal
$Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$	3	3.839	3.839	35.78	530.11	Tetragonal
$Tl_2Ba_2Ca_3Cu_4O_{12+\delta}$	4	3.837	3.837	41.2	615.66	Tetragonal



Figure(1) X-ray diffraction pattern for (Tl-2212).

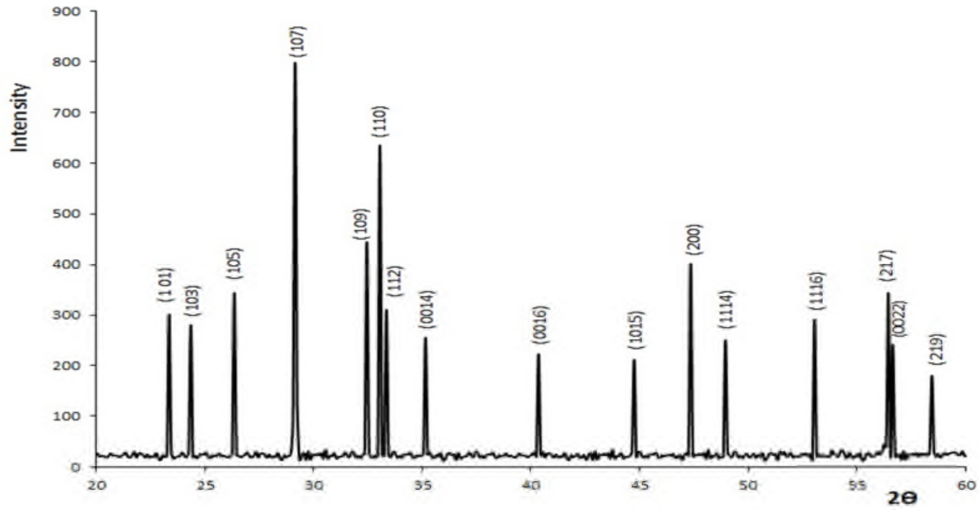


Figure (2) X-ray diffraction pattern for (Tl-2223).

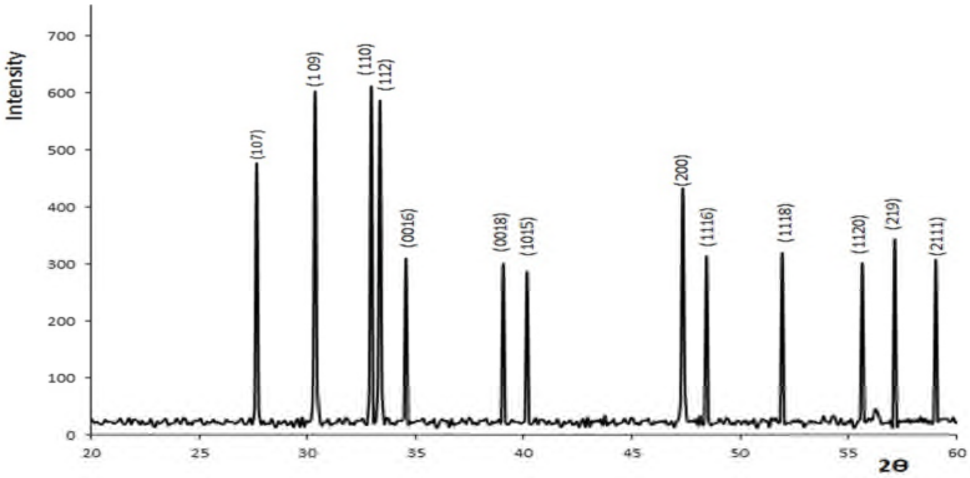


Figure (3) X-ray diffraction pattern for (Tl-2234).

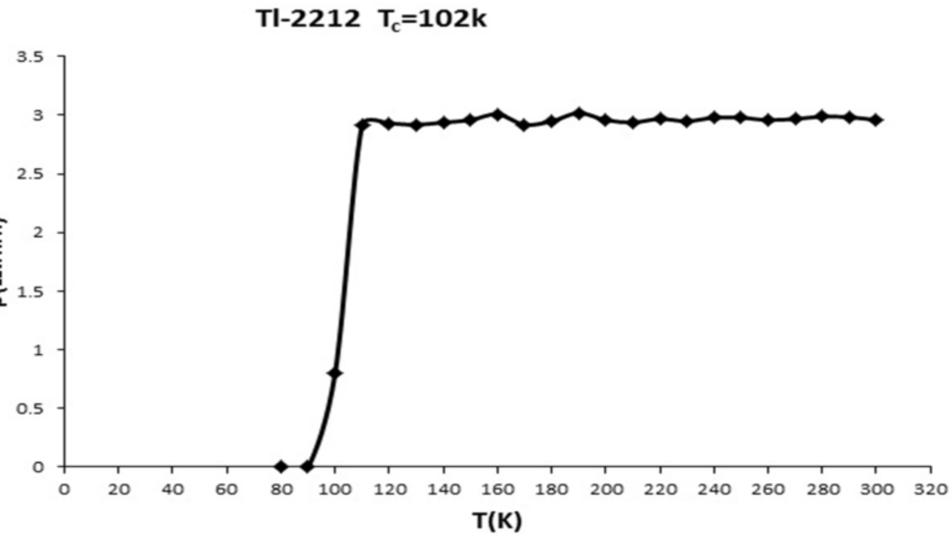


Figure (4) The resistivity behavior versus temperature for prepared $Tl_2Ba_2CaCu_2O_{8+\delta}$

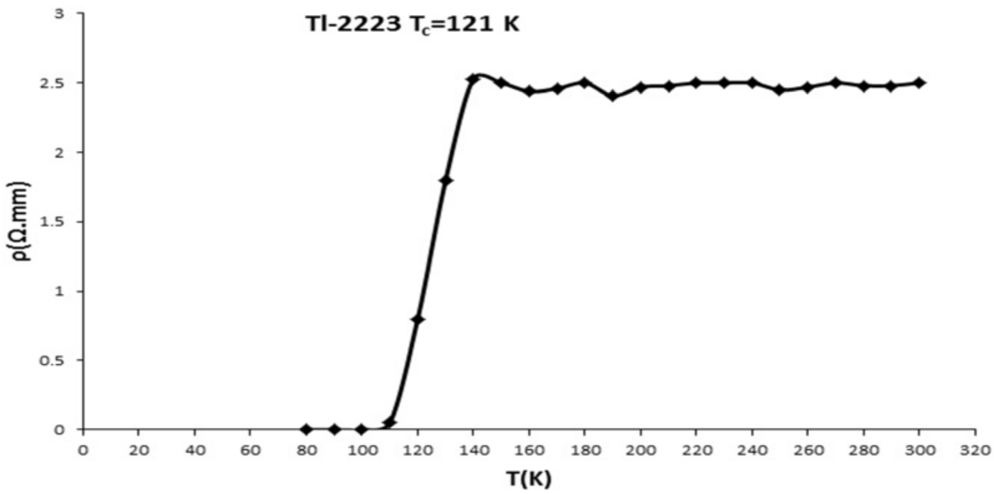


Figure (5) The resistivity behavior versus temperature for prepared $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$

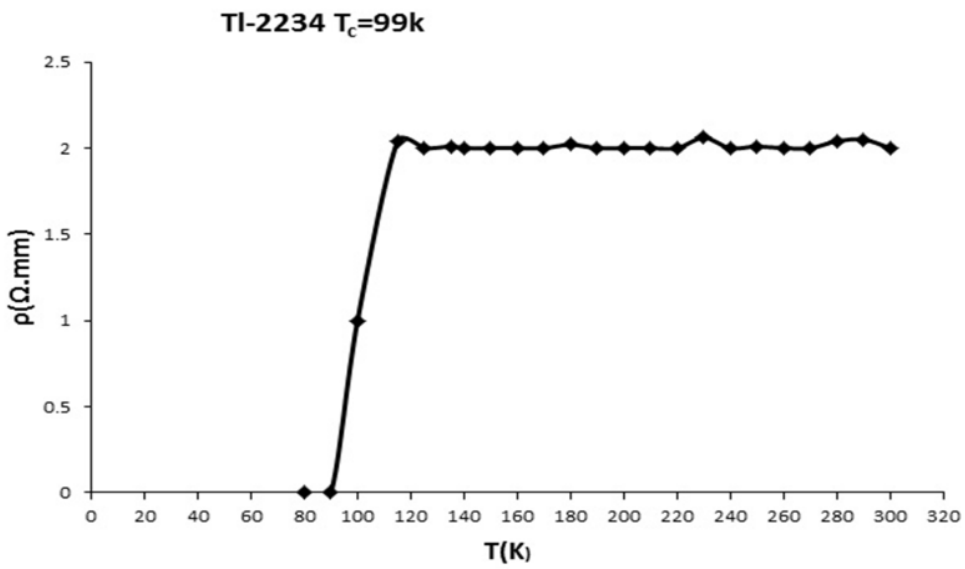


Figure (6) The resistivity behavior versus temperature for prepared $Tl_2Ba_2Ca_3Cu_4O_{12+\delta}$

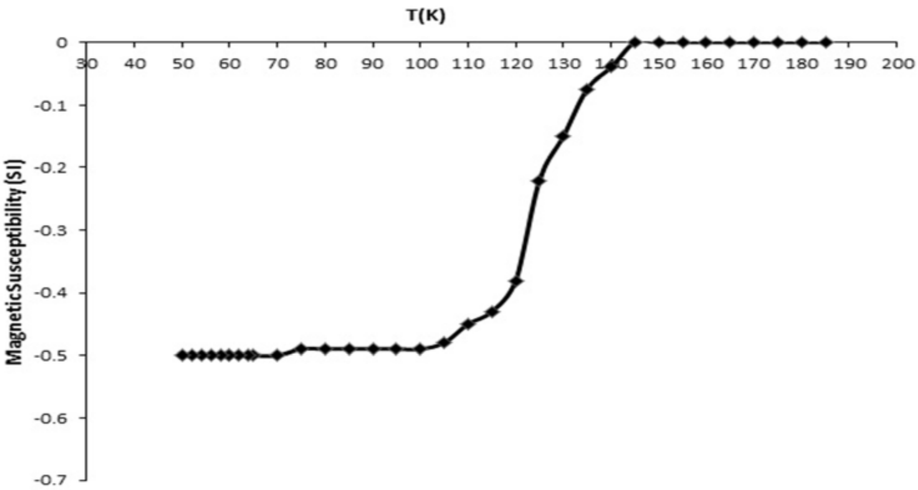


Figure (7) the magnetic susceptibility measurement for the sample of $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$

CONCLUSION

The presence of superconductivity, in the Tl–Ba–Ca–Cu–O system, the phases very sensitive and depend on the sintering temperature. The optimum value of the sintering temperature is 860 ◦C which gives the preferable structure and high T_c. The metallic nature of the Tl–O layers plays an important role in achieving high transition temperatures. T_c is determined by theCuO_2 layer with the highest intrinsic T_c . This is consistent with the large mass anisotropy of the thallium. Highly sensitive magnetic susceptibility measurements on the same crystal as used for structural study yielded a generic parabolic-like T**c** behavior. These layers act as a direct between the Cu–O_2 layers. On the other hand, this layer is considered as a bridge for charge transfer in the *c* direction. The effect of interlayer coupling, the coherence length is shorter than the distance between the two Cu–O_2 planes, so the super-current could then flow between Cu–O_2 layers by taking advantage of the metallic states on the intervening layers, essentially hopping from Cu–O_2 layer to Cu–O_2 layer by tunneling through the metallic interlayer.

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