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Effect of Preparation and Annealing Temperature on the Properties of (Hg,Tl)-2223 Superconductor

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ABSTRACT: *Samples of superconducting compounds were prepared by a solid-state reaction technique in a sealed quartz tube under normal pressure. The impact of the compound on the electrical properties has been studied using the electrical resistance measurements of the samples as a function of temperature. The obtained results appear that an enhancement in the phase formation, and the superconducting transition temperature T_c were improved. It may be due to the decreasing of the magnetic impurities or the delocalisation of carriers which resulted in the enhancement of the density of mobile carriers in the conducting CuO_2 planes.*

Keywords: Superconducting, annealing, temperature resistance zero, mobile carriers

1. INTRODUCTION

Most studies on high-temperature superconductivity (HTS) have been concentrated on reaching the highest superconducting transition temperature, T_c. The homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ [Hg-12(n-1)n] consists of two types of layers, i.e., charge reservoir layers (CRL) $HgBa_2O_x$ and infinite layers (IL) $Ca_{n-1}Cu_nO_{2n}$, where n is the number of CuO_2 planes that exist between two CRLs and supply holes to the above-mentioned CuO_2 planes.[1] Concentration of charge carrier in the CuO_2 planes plays an important role in high T_c superconductors.

T_c of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ phases strongly depend on two parameters: oxygen content (δ) and number (n) of (CuO_2) planes in their structures.[2,3] The number of CuO_2 planes (n) dependence of T_c is an interesting problem that may bring important information to understand the high-T_c superconductivity. Figure 1 shows the dependence of T_c versus n. In this family of materials, the transition temperature sequentially increases with increasing number n of CuO_2 planes up to n = 3, and then it is observed to decrease with further increase of n. Each phase has a different transition temperature to the superconducting state: for n = 1 (1201), T_C = 97 K; n = 2 (1212), T_C = 127 K; n = 3 (1223), T_C = 135 K; n = 4 (1224), T_C = 126 K; n = 5 (1245), T_C = 110 K; and n = 6 (1256), T_C = 107 K.

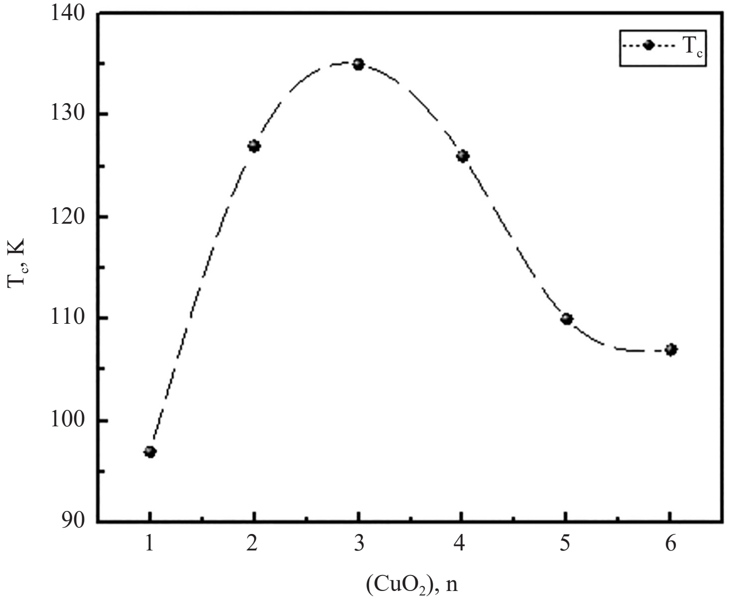

Figure 1: Variation of T_c as a function of n for $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$.

Previous studies on high T_c superconductor have shown that the chemical doping or substation, preparation conditions and hole concentrations play very important roles in high T_c and conventional superconductors.[4,5] To improve critical transition temperature of the (Hg,Tl)-2223 compound, samples were prepared with different conditions of annealing at different temperatures. It is very important to investigate impact of varying annealing conditions on the oxygen content of the (Hg,Tl)-2223 samples. Decreasing oxygen content in the sample leads to increase in T_c. However, if the oxygen content increases, this allows for forming of other phases inside the sample due to the increase of the pressure inside the quartz tube.[6] Thus, it is useful to observe the variation of both the superconducting properties, as well as the normal state properties of materials as a function of changing preparation, annealing temperature and time annealing in order to understand superconductivity better.

Therefore, the aim of the current study is to synthesise and characterise the (Hg,Tl)-2223 high temperature superconductor and to investigate the effects of the preparation, annealing temperature and time annealing on the properties of these superconducting samples. The results of the present study may provide useful information to further studies of the properties of (Hg,Tl)-2223 superconductors and the optimisation of the annealing processes for (Hg,Tl)-2223 superconductors.

2. EXPERIMENTAL

Samples with the nominal composition of $(Hg_{0.1},Tl_{0.9})_2Ba_2Ca_2Cu_3O_{8+\delta}$ were prepared by the standard solid-state reaction method in only one step. High purity (99.95%) chemicals of HgO, Tl_2O_3, BaO_2, CaO and CuO were used as starting materials. These oxides were mixed using an agate mortar to make fine powder which was sieved in 64 μm sieve to obtain a homogeneous mixture. The powder was pressed in discs (1.5 cm in diameter and about 0.3 cm in thickness). Then, these discs were wrapped in a silver foil with 0.1 mm in thickness, which were put in sealed quartz tubes with a diameter of 1.5 cm and length of 15 cm. Next, the sealed tubes were put in closed stainless steel tubes, and the stainless steel tubes were placed horizontally in a furnace. Next, they were heated at a rate of 4°C min^{-1} to (700°C/811°C). The samples have been maintained at this temperature for 6 h, and then they were cooled to room temperature at a rate of 0.5°C min^{-1}. (Hg,Tl)-2223 superconducting samples were annealed in normal atmosphere at 500°C for different time.

A closed cryogenic refrigeration system was used to perform the measurements of DC resistance for all samples with the four-probe method. The four contacts on the samples were made by a conductive silver paint. During resistance measurements a constant current of 2 mA has run through the sample, which was provided from a Keithely 2400 current source to avoid heating effects on the samples.

3. RESULTS AND DISCUSSION

Figure 2 shows behaviour of normalised resistance R (T)/R (300K) versus temperature for (Hg,Tl)-2223 samples before and after annealing. It is clear that the annealed sample has a tail and its resistance zero at a temperature of 79 K. This may be due to missing oxygen content. Figure 1 shows that the annealing has improved the phase transition and the semiconducting-like behaviour in the normal state was absent for the sample S_1. The onset temperature (T_c^{onset}) was increased from 111 K to 117 K. A value of zero electrical resistance (T_c^{offset}) was 63 K before annealing, and became as 79 K after annealing, being higher by the annealing process.

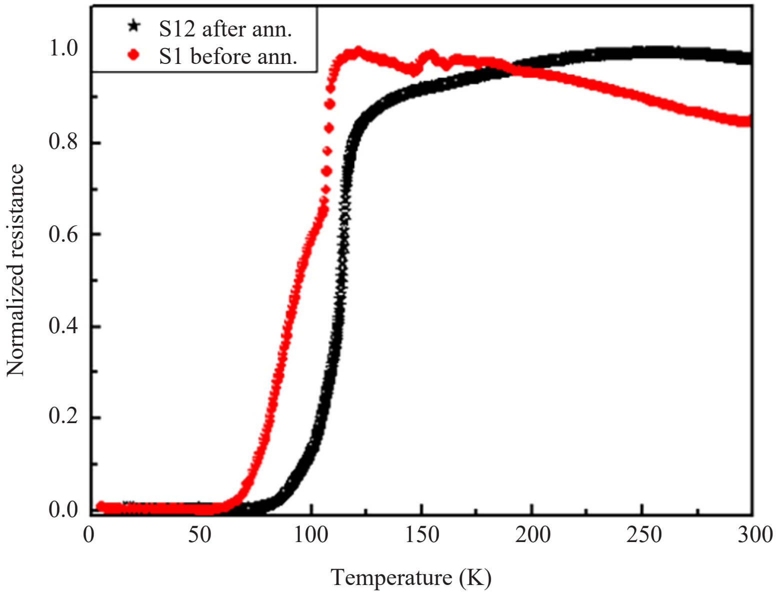

Figure 2: Normalised resistance vs. temperature characteristics for S_1 samples, before (S_1) and after annealing (S_{12}).

There are two possibilities for enhancing T_c in the (Hg,T1)-2223 samples annealed under normal condition. First is by increasing of the amount of oxygen in the bulk to increase the concentration of holes in p-$d_{x^2-y^2}$ bounded. Second is possibility to reduce the amount of extra oxygen from the sample to decrease the undesired phases (magnetic, non-superconductor). It was previously reported that T_c of T1-2223 samples synthesised under ambient pressure was substantially enhanced by the annealing in an evacuated tube.[7]

Figure 3 shows the results of resistance measurements for the as-sintered samples sintered at 700°C (sample S_1) and 811°C (sample S_2) for 6 h. Clearly, it is noticeable that the sample S_1 has two phases, but with the sample S_2, it is almost single phase. This means that the second phase has disappeared when the sample sintered at 811°C. It is important to mention that the S_2 sample was improved in connectivity and the improvement is related to the uniform distribution and alignment of superconducting grains. Also, as shown in Figure 3, the resistance decreases with temperature from 300 K like a metal for the S_2 sample, whereas the S_1 sample does not. The results of R (T) show that the sample has semiconducting-like behaviour in the normal state. This may be due to the reduced oxygen concentration in the bulk that may act as effective channelling centres of oxygen vacancies.[8,9] The values of T_c^{onset} and T_c^{offset}, which are determined from the electrical resistance behaviour are

111 K and 63 K respectively for S_1 sample, while the T_c^{onset} and T_c^{offset} for the S_2 sample are 107 K and 93 K, respectively. The transition width $\Delta Tc = T_c^{onset} - T_c^{offset}$ determined from the difference between the onset temperature and zero resistance temperature for (Hg,Tl)-2223 samples are listed in Table 1.

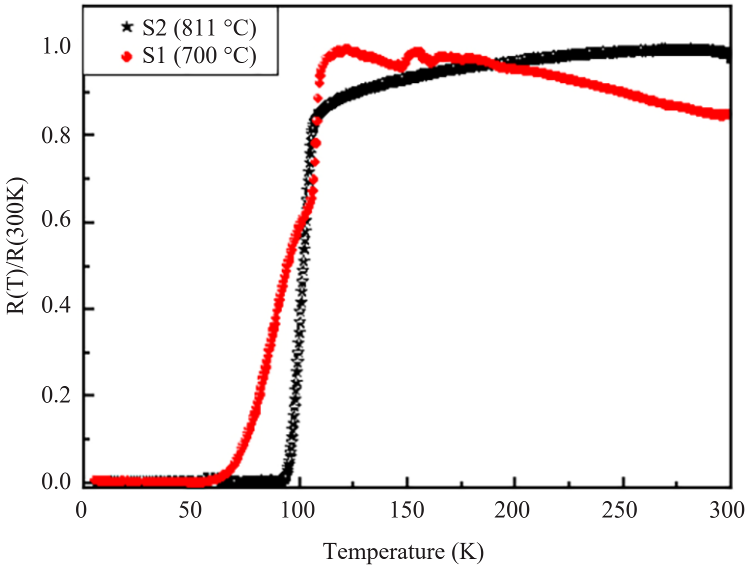

Figure 3: Temperature dependence of the normalised resistance for (Hg,Tl)-2223 sample sintered at 700°C and at 811°C for 6 h.

Figure 4 shows the temperature dependence of a normalised resistance for S_2 samples at different annealing time at 0 T. It is observed that all samples (above the transition temperature onset (T_c^{onset}) have a linear metallic behaviour in the normal state. By decreasing the annealing time, the metallicity increases. More metallic behaviour may be attributed to the best grain connection or to optimal carrier doping in the CuO_2 conducting planes of the sample under high oxygen pressure.[10] T_c^{onset}, determined from the electrical resistance behaviour when the resistance first drops, is equal to 107 K for S_2 sample. When the sample was annealed at 500°C for 4 h (sample S_{21}), T_c increased by about 7 K to 114 K. Further annealing at 500°C for 2 h (sample S_{22}) enhanced T_c up to 122 K. It is clear that the transition width decreased with decreasing annealing time, and the annealing improved the coupling characteristics between superconducting grains. The different values for all samples determined from the resistance measurements are listed in Table 1. Annealing the samples in normal conditions leads to a decrease of resistance and to an increase of T_c, in agreement with the results of other studies.[11–19]

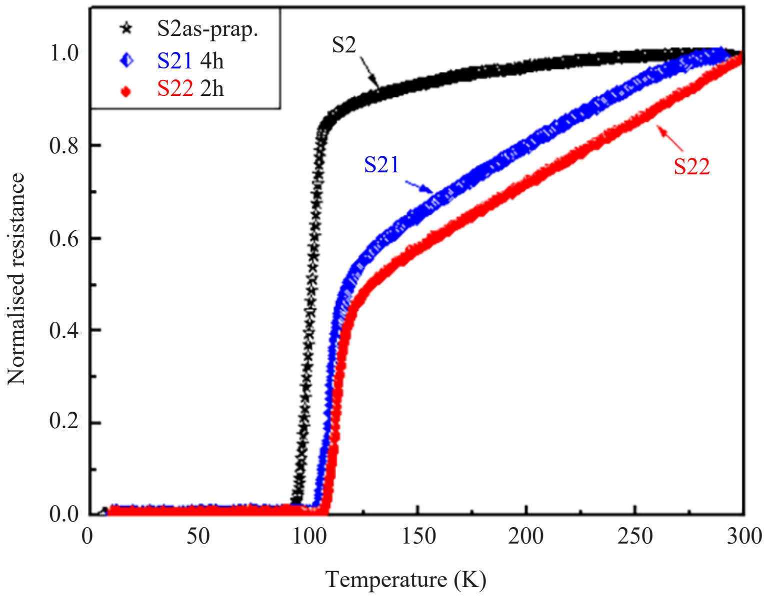

Figure 4: Normalised resistance vs. temperature measurements of S_2 sample annealed at 500°C for 4 h and 2 h.

Table 1: Sample preparation with heat treatment conditions, normal-state resistance (R_{300}), residual resistance (R_0), T_c^{offset}, T_c^{onset}, and ΔT_c from resistance measurement.

Samples	Annealing time (h)	R_{300} (Ω)	R_o (Ω)	T_c^{onset} (K)	T_c^{offset} (K)	ΔT_c (K)
S_1	0	1.7052	2.22875	111	63	48
S_{12}	4	–	3.00446	117	79	38
S_2	0	0.0712	0.06031	107	93	14
S_{21}	4	0.0378	0.00910	114	103	11
S_{22}	2	0.0323	0.00458	122	112	10

The normal-state resistance (R_{300}) and residual resistance (R_0) as a function of annealing time are plotted in Figure 4 for (Hg,Tl)-2223 sample before and after annealing. Here, R_{300} is the resistance at 300 K and R_0 is obtained from the fitting of resistance data in the temperature range $2T_c$ K $\leq T \leq 300$ K, according to Matthiessen's rule.[20]

It can be observed from Figure 5 that the sample S_2 has highest room temperature resistance R_{300} (0.0712 Ω) while the sample S_{22} has lowest R_{300} (0.0323 Ω). The post annealing at 500°C and 811°C (S_{22}) enhances the value of resistance at 300 K. One of the possible contributions to the room temperature resistance is from the defects present in the samples. The defects may be due to the presence of impurities and weak links between the superconducting grains.[21] During annealing, the oxygen content and thus the density of mobile carriers of the superconducting phase could increase, giving rise to a decrease in resistance.[22] After annealing in normal conditions, the S_{22} sample had a critical temperature T_c^{onset} = 122 K with a transition width ΔT_c = 10 K. It may be due to the decreasing of the magnetic impurities or the delocalisation of carriers which results in the increase of the holes concentration in the conducting CuO_2 planes.

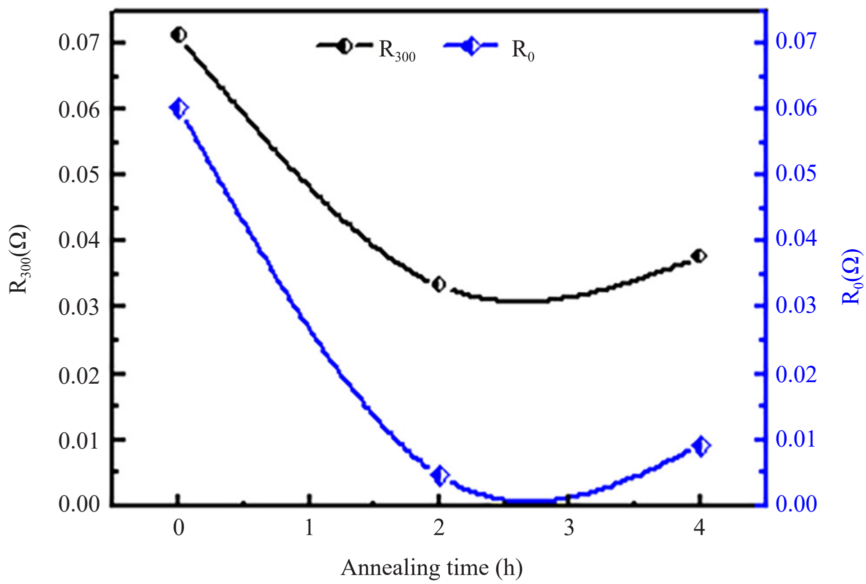

Figure 5: The R_{300} and the R_0 as a function of annealing time for (Hg,Tl)-2223 sample.

4. CONCLUSION

The current study has investigated the influence of preparation, annealing temperature and time annealing on the properties of the (Hg,Tl)-2223 superconductor (T_c^{onset} and T_c^{offset}). The results of R (T) measurements have shown that the optimised annealing temperature is 811°C for 2 h. The highest T_c of S_{22} sample in this study was T^{onset} = 122 K and at T_c^{offset} = 112 K. The increased pressure inside the quartz tube allows the formation of other phases inside the sample when the samples were annealing in air (normal conditions). The inter-grain superconductivity in (Hg,Tl)-2223 samples may be significantly affected by the time and the annealing temperature.

5. ACKNOWLEDGEMENTS

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