

The Correlation of Miller Indices with the Conductivity Mechanism of Cu-Based High- T_c Superconductor

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Abstract

The high temperature superconductor of $\text{CuBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ (Cu-1223) was synthesized experimentally by solid-state reaction. There were many parameters affecting the preparation technique such as oxygen flow, sintering temperature, sintering time, and compacted pressure. The XRD-pattern exhibited a tetragonal phase for both Cu-1223, and Cu-1212. The resistivity measurement was considered the important experiment to show the predominant phase of superconductor if it was a high phase or low phase. The critical temperature (T_c) was about (128 K) for the high phase of Cu-1223, whereas the low phase of (Cu-1212) appeared at ($T_c = 64$ K). The correlation between the Miller indices and resistivity measurements was a good tool to predicate the expected mechanism of conductivity within the unit cell. It was clear that the arrangement of these planes was aligned in the a-c direction. That should make the motion of the charge to be simpler by a stepping plane toward c-axis.

Keywords

Solid-State Reaction, High- T_c Superconductor, Resistivity Measurement, XRD-Pattern, SEM Analysis

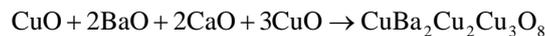
1. Introduction

The high- T_c superconductor cuprate like $\text{HgBa}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{2n+2+\delta}$, with different values of ($n = 1, 2, 3$) was recorded a critical temperature of about ($T_c = 138$ K) at ($n = 3$) [1]. This compound was classified as Hg-family, and the most interesting focused upon the third member ($n = 3$) by the series (Hg-1223) due to its high critical temperature. Many attempts were made to develop Hg-family high- T_c superconductor; part of them were applied the preparation methods,

physical or chemical methods [2] [3]. The other attempts concentrated on the preparation condition of the applied method [4]. The last group depends on the partial substitution of isovalent atoms [5] [6], and the change the parameter ($n > 3$) [7] [8] [9]. It was difficult to obtain the single phase of Hg-family ceramic compound because it contains both high phase Hg-1223 and low phase (Hg-1212). The most important thing in production of Hg-family high- T_c superconductor was how to control the oxygen pressure and mercury pressure; for this reason there were no specific values for oxygen and mercury pressures [10]. The normal solid-state reaction was not used in preparation of Hg-family superconductor because of high evaporation of Hg-element in the mixture; to avoid this problem, the technique of a sealed quartz tube applied thermally in solid-state reaction [11]. The solving of this problem was the replacing of a metal ion instead of Hg-ion such as Cu-ion, and producing a new family like Hg-1223 family [12] [13]. That was the aim of the research, and studying the electrical and structural properties of Cu-1223 was the main purpose to specify this superconductor type like Hg-1223. The correlation between them should be important to understand the conductivity mechanism, as shown in **Figure 1**. The previous studies appeared the multi-phase in (Hg-1223) compound, and then the multiphase is acceptable in Cu-1223.

2. Experimental Work

The Cu-1223 compound synthesized by solid-state reaction using the principle materials like CuO, BaO, CaO, with an appropriate weight in proportion to their molecular weights according to this reaction.



The ceramic sample of $\text{CuBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ calcined in air at (850°C) for (24 h) with a heating rate (45°C/hr) and cooling rate (40°C/hr). Then the sample gets

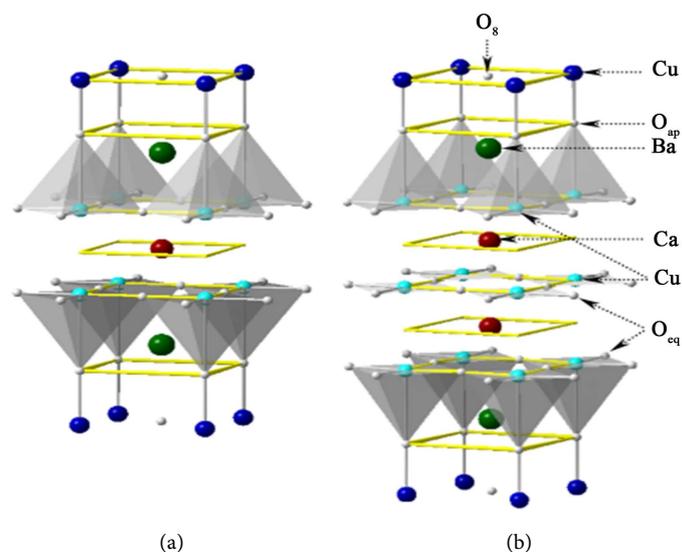


Figure 1. The unit cell of superconductor compound (a) Cu-1212 and (b) Cu-1223 families.

into a pellet form with a diameter (1.5 cm) and thickness (3 cm). The pelletized sample sintered at a temperature (900°C for 24 h) in oxygen flow by a rate of about (0.5 L/m) under a heating rate (45°C/hr) and cooling rate (40°C/hr). Later, there were many samples examined by XRD analysis and resistivity measurement in order to make a correlation between them. This correlation would help us to understand the conductivity mechanism in Cu-1223 high temperature superconductor.

3. Results and Discussion

The prepared sample of $\text{CuBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$ compound studied by the X-ray diffraction to find the structural nature and the presence of single or multiphase within the structure. The XRD diffraction pattern for Cu-1223 had appeared as in **Figure 2**.

The obtained results showed the peaks related to high superconductor phase Cu-1223, which were more than the low phase of Cu-1212. The ratio of high phase was about 74% of the whole structure. This result agreed with the data sheet concluded from database information for low phase Cu-1212 and high phase Cu-1223. The appearance of two phases accepted due to a polycrystalline structure. The XRD analysis was applied in the Refine program in order to find the lattice parameter $a = b = 4.171 \text{ \AA}$ and $c = 16.743 \text{ \AA}$. It found that Cu-1223 had a tetragonal phase, and the lattice constants were larger than the lattice parameter for Hg-1223 especially in the basal plane [8]. That was might be attributed to high oxidation of Cu-ion rather than Hg-ion in the basal plane, which made more absorption of extra oxygen in comparison with Hg-1223. The database of (ICDD) with pdf file (98-004-1652) was used to analysis the pattern of Cu-1223 system. The small change in lattice constant might be related to the variation of (2θ) position. The observed change in lattice parameters was agreed with the strain happened in Cu-1223 compound that was about (0.885 ± 0.149) .

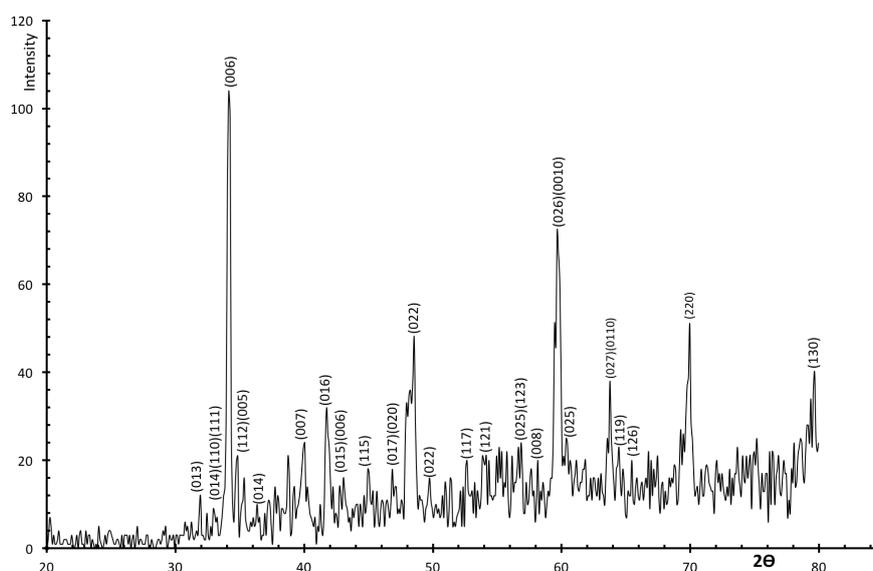


Figure 2. The XRD-pattern of the compound $\text{CuBa}_2\text{Ca}_2\text{Cu}_3\text{O}_{8+\delta}$.

The planes of low phase had peaks (013), (005), (014), (015), (006), (022), and (025) with different intensity at ($2\theta = 31.951, 35.349, 36.356, 42.758, 43.053, 49.761, \text{ and } 60.417$) respectively. They were compared with the international center for diffraction data (ICDD) with pdf file (98-007-4104), it was found that the planes (006), (007), (008), (0010) at ($2\theta = 34.153, 40.062, 58.178, \text{ and } 59.704$) had high intensity in Cu-1223 rather than Cu-1212. The full Cu-substitution appeared by the replacing Cu-ion instead of Hg-ion in the basal plane as mentioned in **Figure 1**. It found the plane (006) had the same value of 2θ in Hg-1223. Whereas the plane (007) had a small shifting in 2θ . On the other hand, the plane (0010) created in Cu-1223. These planes considered a conducting plane because they related to charges connection through the c-axis.

The planes (110), (130) and (220) in Cu-1223 had the same effect of the plane (110) in Hg-1223 compound. They linked the basal planes of both the bottom and the top of unit cell. On the other hand, the plane (020) was making the correlation between both basal planes in the unit cell. It was indicated the presence of extra oxygen in the basal planes of Cu-1223 unit cell. As well known, the conductivity mechanism depended on the motion of the charge carriers through the principle planes along the a-c direction. That means the planes (022), (026), (027), (014), (016), (017), (0110), and (025) considered the stepping layers for the charge motion toward c-axis during the chain. There were two common peaks (115) and (111) of Cu-1223 and Hg-1223 but at different intensity and a small shifting in 2θ .

The appearance of new planes (112), (117), (121), (123), (119), and (126) at ($2\theta = 34.855, 52.646, 54.045, 65.889, 64.453 \text{ and } 65.47$) in Cu-1223 compound. These planes acted as a correlation factor between the planes along the c-axis. Increasing the concentration of these planes tend to increase the factors affected the mechanism of conductivity and then improve the T_c -value. Then the enhancement of superconductivity mechanism was happening during the presence of Cu-ion in the basal plane and forming the active bond (Cu_2O) linking the basal plane. The common thing was the appearance of Cu-1223 phase like Hg-1223 with some variation in the lattice constant. This result might be a positive effect on the resistivity measurement in order to determine the superconducting phase as mentioned before [11]. The small difference in the crystal structure related to the appearance of low phase Cu-1212, which formed by the ratio 26% of the whole structure. The improvement of the physical properties of Cu-1223 compound done by removing the low phase Cu-1212. The pre-sintering of the sample under study examined by the XRD pattern as shown in **Figure 3**.

It was found that the high phase Cu-1223 formalizes 45% of the whole structure and the low phase Cu-1212 formalize 55% of the whole structure, so the low phase Cu-1212 was the predominate phase. The common peaks between the two last figures had the planes (005), (022). The first plane had a small shifting in (2θ) and the second had the same position of Hg-1212 phase. There were a common planes for Cu-1223 and Hg-1223, such as (003) at ($2\theta = 21.361$) for Cu-1223 and it was appeared at ($2\theta = 21.56$) for Hg-1223. The planes (022),

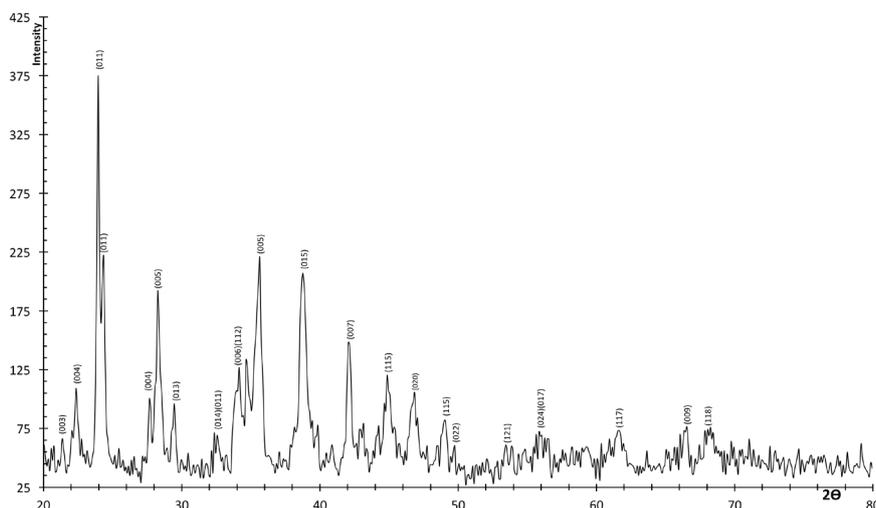


Figure 3. The XRD-pattern of the compound $\text{CuBa}_2\text{Ca}_1\text{Cu}_2\text{O}_{8+\delta}$.

(011), (024) and (017) were represented the stepping layers to satisfy the conductivity mechanism during the motion of charge carriers along the a-c direction through principle planes (005), (003), (004) and (009) as mentioned a conducting layers along c-axis. It observed that the appearance of those planes in Cu-1212 by the pre-sintering made less than in the first one. The plane (020) had played the same role in the high phase. It linked the basal plane and investigated the structural symmetry. There was a new plane (118) occurred in the low phase. The applying of Refine program tend to conclude the lattice constant $a = b = 3.55 \text{ \AA}$ and $c = 12.37 \text{ \AA}$ as a tetragonal phase.

The results showed that there was a small difference from the (ICDD) pdf number (98-007-4104), by the change in 2θ -position of the planes. They were agreed with the result of the strain in the Cu-1212 that was about (0.314 ± 0.194) . One can conclude that the pre-sintering was the reason to predominate the low phase of Cu-1212. In general, the carrier motion directed in the a-c direction for Cu-1223 and Cu-1212 as mentioned before [14]. The small difference in the lattice constants between them related to the extra oxygen, which pumped during the pre-sintering process. It was interested to note that the conducting planes represented by (006), (007), (0010), (008) at ($2\theta = 34.153, 40.062, 59.704, 58.178$) respectively. Moreover, the plane (006) had a high concentration in the crystal structure, which was the reason to form (O-O) bond. The mechanism of conductivity must be done during the motion of charge carriers through the stepping planes along the a-c direction represented by the planes (014), (016), (017), (022), (025), (026), (027), and (0110) at ($2\theta = 32.462, 41.748, 46.854, 48.549, 56.652, 59.501, 63.571, 63.74$) respectively. It could be seen that the previous planes contributed to achieve the structural symmetry by linking both basal planes represented by (110), (220), (130) at ($2\theta = 32.978, 69.984, \text{ and } 79.647$) respectively. The excess of oxygen atoms were the way to connect the basal planes (020) at ($2\theta = 47.137$). It also contributed to achieve the symmetry in the structure. There were other planes related to high phase of (Cu-1223) such as (111), (112), (115), (117), (121), (123), (119), and (126) at ($2\theta = 33.249, 34.855,$

44.958, 52.646, 54.045, 56.889, 64.453, and 65.47). On the other hand, the planes were mentioned by (013), (005), (014), (015), (006), (022), and (025) at ($2\theta = 31.951, 35.349, 36.356, 42.758, 43.053, 49.761, \text{ and } 60.417$) related to the low phase of (Cu-1212).

The electrical resistivity was the most important factor to show the superconductivity, and it considered a good tool to show the correlation of miller indices with interlayer coupling of the charge carriers. The last was the reason for understanding the conductivity mechanism. The resistivity measurement done by using a closed cycle cryogenic system worked by liquid helium, as shown by **Figure 4** for Cu-1223 under the first sintering. It was clear that there were two regions in the resistivity curve one of them belong to high phase of Cu-1223 and the other belongs the low phase of Cu-1212. A predominate of high phase Cu-1223 was exhibited a normal resistivity of about $0.0215 \Omega \cdot \text{mm}$, and critical temperature was about 128 K. The low phase of Cu-1212 showed normal resistivity of about $0.0115 \Omega \cdot \text{mm}$, and a critical temperature of about 66 K. The appearance of the two phases and the predominated high phase of Cu-1223 was supported the result of XRD during the high concentration of the planes including in Cu-1223. There were many peaks related to Cu-1223 such as (006), (007), (008) and (0010) as mentioned before. These planes are responsible on the conductivity mechanism because they considered correlated planes within the unit cell. They considered linking planes for charge transfer. The presence of two phases, low and high phase, is confirmed by Ginzburg-Landau theory [15]. They stated the presence of two phases during different temperature range and they said there was a vortex state including the low and high phase superconductor. That was defined a type-II high temperature superconductor.

The pre-sintering had been done to explain the more oxygen excess that was the reason to conform the low phase as shown in **Figure 5**. That related the

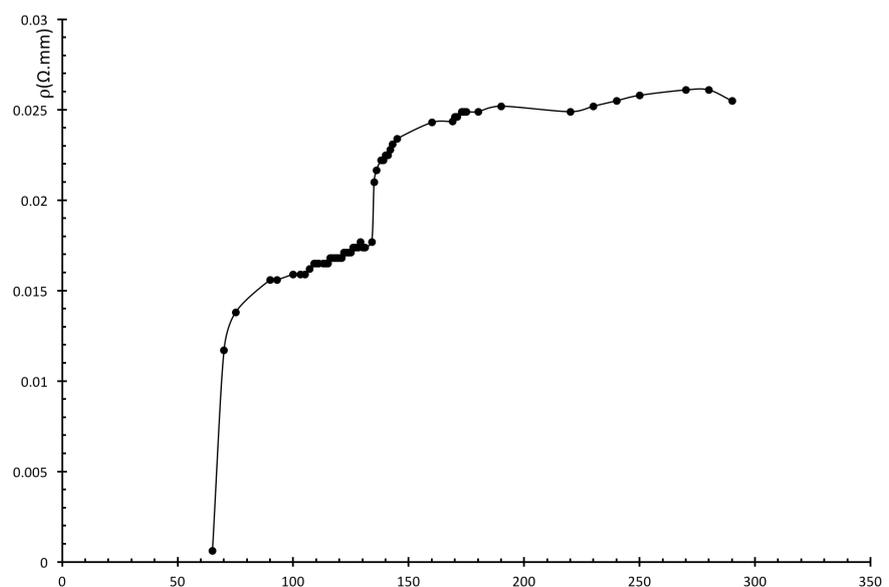


Figure 4. Resistivity measurement for the sintered Cu-1223.

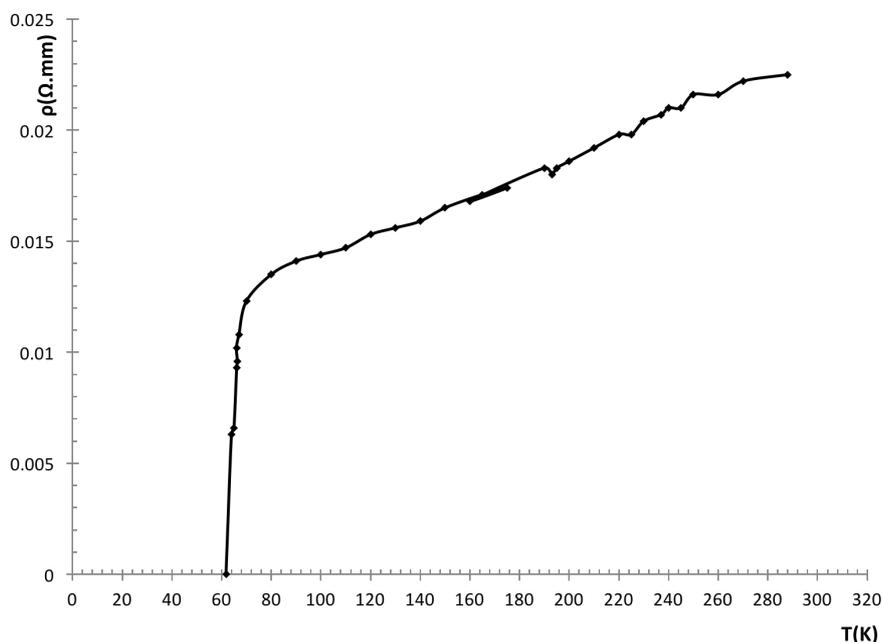


Figure 5. Resistivity measurement for the pre-sintered of Cu-1212.

complete occupation of the vacant site in the basal planes, which was the reason to restrict the motion of charge carriers through the stepping planes in order to link the basal planes. The resistivity measurement showed the single low phase of Cu-1212, it supported by XRD where the low phase predominated within the crystal structure. The critical temperature was about 64 K, then it was interested to note that sample after the pre-sintering the normal resistivity had 0.01 Ω-mm, half the normal resistivity of the sample before the pre-sintering. That related to the presence of a single phase rather than the multiphase for the sample prepared after the first sintering. On the other hand, the pre-sintering means the long sintering time and the absorption of extra oxygen was more increased, which had a direct effect on the concentration of charge carriers producing a high conductivity. Then the pore size distribution decreased and the sample became dense, that was the reason why the conductivity increased after pre-sintering. It observed was noticed, the T_c -value had half the T_c -value of the sample before pre-sintering that was related to diminish the most peaks causing the high phase Cu-1223.

4. Conclusion

The producing of new high T_c superconductor compound like Hg-1223 phase is possible by the replacement of Cu-ion by Hg-ion in the basal plane. A high phase of Cu-1223 created a tetragonal structure through XRD spectrum, which was different from the phase Hg-1223. There were more Miller planes in Cu-1223 those were the reason to enhance the conductivity mechanism. The motion of charge carriers was investigated during the a-c direction, toward the c-axis. On the other hand, there were both phases created, high phase Cu-1223 and low phase Cu1212 but the predominant one was Cu-1223. The pre-sintering

done to show the single phase was present, but the conductivity mechanism was continuing through a-c direction forward to c-direction. These results considered a good tool to understand the conductivity mechanism. It considered a base on the future work related. The applying of these results was theoretically the main future topic to give a complete view on the conductivity mechanism in high temperature superconductor like Cu-1223 phase.

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