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Disordered continuum percolation in YBCO/CuO composites

D R Mishra

***Abstract*— High T_c superconducting systems namely BSCCO and YBCO were studied for the modes of percolation process. The study was carried out on a series of specimens of bulk polycrystalline superconducting-insulating composites prepared in CuO matrix. The specimens of BSCCO and YBCO system were prepared as $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_\delta$ + x% CuO (namely BPSCCO+CuO) series and $Y_1Ba_2Cu_3O_{7-\delta}$ + y% CuO (namely YBCO+CuO) series respectively, with x and y values in the range of 0-100% (by weight). The lattice percolation model and the continuum model fit to the observed data. It was observed that BSCCO and YBCO system comply to the lattice percolation model and the continuum model respectively. It is because the conduction in BSCCO system takes place through a well-defined Cu-O lattice and the added CuO grows in continuation to this already existing lattice. On the other hand, the addition of matrix in YBCO system creates a continuous conduction medium very different from the spread lattice conduction sites. The conduction in this system needs Cu-O chains, which do not develop with the added Cu-O matrix.**

***Index Terms*— Cuprate superconductors, electrical resistivity, percolation effects.**

I. INTRODUCTION

It has been a difficult task to correlate the conduction process in high T_c superconductors with the structure and arrangement of atoms in the three-dimensional lattice of these materials. One of the ways to look at this problem is by studying the percolation of conduction electrons through a matrix of high T_c superconducting grains embedded in an insulating material. According to the random disordered continuum model [1], the charge conduction in the bulk polycrystalline superconducting-insulating composites is through the holes of zero conductivity which are considered as dispersed within a three-dimensional continuum sheet. The flow of the conducting fluid through such constricted path is studied and the percolation behavior is evaluated. On the other hand, according to the standard three-dimensional lattice percolation model [2], there are conducting sites connected to each other through bonds which provide path for the conducting fluid (here, composed of superelectrons). The conduction is either through “site-percolation modelling” where each site in the lattice has a finite probability of occupancy and packing factor (depending on the symmetry) or through “bond $^{\text{percolation}}$ modelling” where there is a critical number of bonds per site for the percolation of conducting fluid. A minimum superconducting volume fraction (SVF) called the percolation threshold (P_c) or an inter-particle separation smaller or comparable to the coherence length, is required for the resistive superconducting transition in High T_c materials [3]. The percolation threshold for the thermoelectric superconducting transition in the bulk polycrystalline superconducting-insulating composites is lower as compared to the percolation threshold for the resistive superconducting transition in these materials [4,5].

A. Mammou et al [6] have shown that $Y_1Ba_2Cu_3O_{7-\square}$ +CuO binary mixture shows a percolation threshold for resistivity corresponding to a superconducting volume fraction of 23%. They also showed that critical current density J_c follows a power law of the form $J_c=J_{c0}(p-p_c)^v$, where p is the superconducting volume fraction and p_c is the threshold value. J J Lin et al [7] studied percolation behaviours of a BSCCO +Al_2O_3 mixture. They found that disordered continuum model [1] is applicable to the mixture.

These reports however, did not discuss the reasons why a particular superconductor-insulator mixture complies with a particular model viz. the lattice percolation model vis-a-vis the continuum percolation model. Therefore, it was decided (by us) to investigate the binary mixtures $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_\square$ +CuO and $Y_1Ba_2Cu_3O_{7-\square}$ + CuO, in order to get a correlation with above theoretical models.

This paper reports the difference between the percolation behaviors of BPSCCO + CuO and YBCO + CuO composites which is because of the difference in the conduction mechanism in the two composites. BPSCCO + CuO mixture has conducting Cu-O planes whereas YBCO system does not have such well–defined Cu-O conducting planes and requires both plane and chain for the conduction to take place.

II. EXPERIMENTAL

Superconducting samples of $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_\delta$ and $Y_1Ba_2Cu_3O_{7-\delta}$, were prepared using the standard solid state-ceramic technique. (BiPb)-2223 samples, after repeated grinding and calcination, were sintered at 830°C for 72 hr in air. $Y_1Ba_2Cu_3O_{7-\delta}$ samples were calcined at 850°C for 24hrs, crushed, ground and then recalcined at 875°C for 24 hrs, mixed and ground again. Copper oxide (CuO) in desired proportion, by weight, was mixed with the two superconducting powders to prepare the two series of composites. These are $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_\delta$ + x% CuO (namely BPSCCO+CuO) series with x = 0, 20, 40, 60, 80 and $Y_1Ba_2Cu_3O_{7-\delta}$ + y% CuO (namely YBCO+CuO) series with y = 0, 20, 40, 70, 75 and 80. The mixtures were ground, palletised and finally sintered at 830°C for 72 hr in air for BPSCCO+CuO and at 920°C for 24hrs under flowing oxygen for YBCO+CuO series of specimens.

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D R Mishra, R.H.Government Post Graduate College, Kashipur, U.S.Nagar, Uttarakhand , India

The resistivity vs. temperature (R-T) measurements were carried out using the standard four-probe method. The estimated accuracy for the resistivity measurement is $\pm 10^{-8}$ Ohm-cm. A calibrated Si-diode, DT-500 (Lake Shore) was used for measuring the specimen temperature. The accuracy for temperature measurements is ± 0.01K. The crystal structures of these materials were investigated by obtaining room temperature X-ray diffraction patterns of the specimens on a Sieman's D-500 powder X-ray diffractometer using CuK_{α} radiation. The grain morphology of most of the specimens was studied using the scanning electron microscopy (SEM) technique.

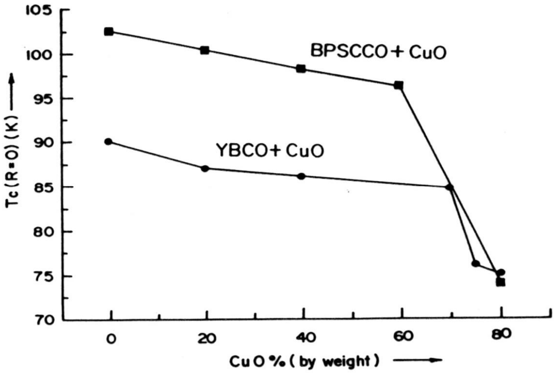

Fig.1. Transition temperature $T_c(R=0)$ vs. CuO concentration (% by weight) of $Bi_{1.6}Pb_{0.4}\mathbf{Sr_2Ca_2Cu_3O}_{\delta}$ + CuO and $Y_1Ba_2Cu_3O_{7-\square}$ + CuO mixtures

III. RESULTS AND DISCUSSION

T_c(onset), $T_c(R=0)$ values and X-ray analysis

Specimens of both BPSCCO+CuO and YBCO+CuO series with CuO concentration less than 85% show superconducting transition. The onset of superconductivity, indicated by a change in slope of the resistivity vs temperature curve is observed at a temperature, called as superconductivity transition onset temperature or T_c(onset). The T_c(onset) =109.8K for pure BPSCCO specimen is higher than the T_c(onset) = 109.6K for BPSCCO+20% CuO specimen. However, the variation in T_c(onset) values for BPSCCO+CuO specimens is very small (≤1K) and T_c(onset) values lie in the range of 109K-110K. The observations were similar in the case of YBCO+CuO series of specimens. Pure YBCO specimen has T_c(onset) ~ 91K and other specimens of YBCO+CuO series have T_c(onset) values in the range of 90-91K.

Fig.1. shows the transition temperature $T_c(R=0)$ corresponding to zero resistivity of the material, plotted as a function of CuO concentration for both BPSCCO+CuO and YBCO+CuO mixtures. The transition temperature $T_c(R=0)$, as shown in the Fig.1 decreases with an increase in the CuO concentration. The transition width $\Delta T = T_c(\text{onset}) - T_c(R=0)$ for both the series of specimens, however, increases with an increase in the CuO concentration. The transition temperatures $T_c(R=0)$ for specimens of BPSCCO+CuO and YBCO+CuO series with 80% CuO are 74K and 75K respectively. The specimens with the concentration of CuO higher than 80% show the onset of superconductivity but do not show zero resistivity down to 65K. Therefore, the phenomenon of percolation i.e. the tunneling of superelectrons through the superconductor-insulator matrix is observable in both YBCO+CuO and BPSCCO+CuO series for CuO concentrations more than 80%.

Fig.1. also shows that $T_c(R=0)$ for specimens with less than 60% CuO in BPSCCO+CuO series and with less than 70% CuO in YBCO+CuO series lie on a nearly straight line curve running at a small angle with the CuO concentration axis. This indicates the existence of an upper limit for the CuO concentration in these mixtures below which the transition temperature $T_c(R=0)$ is close to the transition temperature of the pure specimen. The specimens with higher CuO concentrations show $T_c(R=0)$ very different and very low as compared to the transition temperature of the pure and the other specimens. These observations suggest drastic deformations and structural modifications occur in these specimens with very high CuO concentrations.

The analysis of XRD patterns as discussed in detail, in our previous article [8] on these series of specimens show the peaks corresponding to pure BPSCCO or YBCO phase, the peaks corresponding to pure CuO material, and the peaks corresponding to the modified d- or theta- values. The peaks corresponding to pure CuO material and corresponding to modified d- or theta – values increase with an increase in the CuO concentration. It indicates that the deformations and structural modifications increase with an increase in the CuO concentration. The SEM pictures of BPSCCO + CuO specimens [8] show typical platelet type (BiPb)-2223 particles, which decrease in number with increasing CuO concentrations, only confirming the XRD observations. Specimen with 80% of CuO concentration shows appreciable number of (BiPb)-2223 grains. The SEM observations for YBCO specimens are similar. The observations regarding T_c(onset), $T_c(R=0)$, X-ray diffraction and SEM pictures, however do not help in developing any fresh insight into the conduction mechanisms through these composites. The analysis of the computer fit to these results, however, shows striking dissimilarities in the modes of conduction of superelectrons through BPSCCO + CuO and YBCO + CuO series of specimens. These results and the conclusions drawn are discussed below.

IV. EVALUATION OF THE PERCOLATION THRESHOLD P_C AND 'T' VALUES

The conductivity (σ) of a composite with a conducting fluid flowing through an insulating matrix near the percolation threshold [2] is related to the CuO concentration by the following expression

$$\sigma = \sigma'(P-P_c)^t \quad \ldots(1)$$

where 't' is an exponent which depends on the dimensionality of the conduction network and constant σ' is defined as the conductivity for $P-P_c = 1$. P is the percentage concentration, by weight, of the superconducting material (BPSCCO or YBCO) in the mixture and is fixed for a particular specimen. The threshold or P_c value is the minimum value of the percentage concentration, by weight, of the superconducting material (BPSCCO or YBCO) in the mixture for which the resistive superconducting transition is observed. It is fixed for a particular series (BPSCCO + CuO or YBCO + CuO) of specimens. It was evaluated graphically through computer simulations as described below.

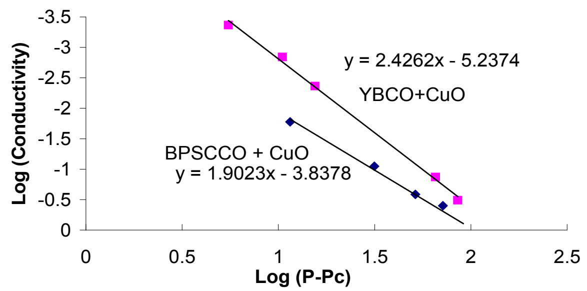

Fig.2. Linear fit to curve showing log of conductivity of $Bi_{1.6}Pb_{0.4}\mathbf{Sr_2Ca_2Cu_3O_\delta}$ + CuO and of $Y_1Ba_2Cu_3O_{7-□}$ + CuO mixtures plotted against the log(P-P_c) at 120K, P being the concentration of the superconductor in the mixture and P_c is threshold value.

Different P_c values, around 20% were chosen and using a graphic software (for example, Microsoft Excel) Log (σ), the logarithmic conductivity was plotted against Log(P- P_c) for a fixed temperature. A straight line fits the experimental data to equation (1) at that temperature.

The best-fit straight lines for BPSCCO+CuO specimens were obtained for $P_c = 0.085$. A representative straight-line plot of Log(conductivity) vs. Log (P-P_c) at T=120K for BPSCCO+CuO and YBCO+CuO series is shown in Fig.2. This P_c value of 0.085 corresponds to the concentration of the superconducting phase ~8.5% (by weight) and is, therefore, the percolation threshold for the BPSCCO+CuO specimens. However, for YBCO+CuO specimens, higher percolation threshold of 14.5% ($P_c = 0.145$) was evaluated by successive fitting of the equation (1) to the experimental data. The reason for this low value of percolation threshold for BPSCCO+CuO series is the existence of well-defined CuO planes in BPSCCO lattice which play an important role in the conduction of the superelectrons through the lattice. The addition of CuO increases the number of CuO planes and is supportive to the conduction mechanism. This continues up to a low concentration of the superconducting phase (~8.5%). If the CuO concentration is further increased, it leads to the distortion of the lattice and finally the superconductivity is destroyed. However, the conduction in YBCO specimens is primarily through Cu-O chains and the role of CuO planes in conduction through YBCO+CuO specimens is not very significant. Therefore, the percolation threshold for YBCO+CuO specimens (~14.5%) is higher comparable to the percolation threshold for BPSCCO+CuO specimens (~8.5%). This difference in the conduction mechanisms in BPSCCO+CuO and YBCO+CuO specimens is further reflected in the analysis of the evaluated exponent t values in equation (1) for the two series of specimens, as discussed in the following section.

The value of the exponent't' corresponding to a particular temperature is evaluated by measuring the slope of the straight line fit to Log (Conductivity) Vs Log (P-P_c) plot at that temperature. The representative linear plots of Log(conductivity) Vs Log (P-P_c) at T=120K (shown in Fig.2) yield t=1.9023 and 2.4262 for BPSCCO+CuO and YBCO+CuO series respectively. Similar plots of Log(conductivity) Vs Log (P-P_c) at different temperatures were drawn and the values of the exponent 't' for both the series were evaluated. Table I shows the values of exponent 't' at different temperatures for BPSCCO+CuO and YBCO+CuO series.

Table 1: The values of exponent 't' at different temperatures for BPSCCO+CuO and YBCO+CuO series.

Serial No.	**BPSCCO specimens**		**YBCO specimens**	
	Temperature in Kelvin	Exponent 't'	Temperature in Kelvin	Exponent 't'
1.	120	1.9023	100	2.3995
2.	140	1.8950	120	2.4262
3.	160	1.8835	140	2.4032
4.	180	1.8788	160	2.3799
5.	200	1.8739	180	2.3582
6.	220	1.8790	200	2.3358
7.	240	1.8833	220	2.3133
8.	280	1.8807	240	2.3078

V. FIT OF THEORETICAL PERCOLATION MODELS

According to the standard lattice percolation model [2] for dimension d≥3, the exponent't' of Equation.(1) is given by

$$t= (d-2)\nu + 1 \qquad ....(2)$$

where ν is the critical exponent of the percolation correlation length. For d = 3 and ν = 0.88 (which is observed in 3D model), t = 1.88. On the other hand, according to the continuum model [1],

$$t = (d - 2)\nu + d - 3/2 \qquad .. (3)$$

and for the same set of values of ν and d, t = 2.38. A comparison of these theoretically predicted 't' values with our experimental results (see Table I) shows that the 't' values for BPSCCO+CuO mixture are closer to those predicted by Equation (2) and therefore, are in accordance with the standard lattice percolation model. On the other hand, the 't' values for YBCO+CuO mixture are closer to those predicted by Equation (3) and therefore, are in accordance with the continuum model.

It may be conjectured that since the BPSCCO lattice consists of well-defined conducting Cu-O planes. The addition of CuO matrix to BPSCCO phase, further, leads to the formation of more of such conducting Cu-O planes and the percolation of superelectrons, through the lattice, is not affected. However, it is noteworthy that in BPSCCO+Al_2O_3 mixture, a higher critical exponent 't' has been reported [7]. The difference is expected, as Al-O planes cannot substitute for the Cu-O planes in BPSCCO lattice and percolation of conducting superelectrons will be through a modified and distorted lattice.

However, in case of YBCO, it is well known that there are no such well-defined conducting CuO planes and both CuO plane and O-Cu-O chains are involved in the conduction of the superelectrons. Hence, YBCO+CuO mixture does not comply with the lattice percolation picture. Presence of distinct phases of YBCO and CuO within the overall matrix makes the system amenable to the continuum model.

CONCLUSION

Low value of percolation threshold ~8.5% and 't' values close to 1.88 for BPSCCO+CuO series indicates that the percolation in the BPSCCO+CuO mixture follows the lattice percolation model. This is because of the development of conducting Cu-O planes within the basic CuO matrix and the percolation of the superelectrons is affected. YBCO system, on the other hand, does not have such well-defined Cu-O conducting planes and it requires both plane and chain for conduction to take place. Therefore YBCO+CuO series follows disordered continuum percolation model and has a higher value of the percolation threshold ~14.5%. The 't' values close to 2.38 for YBCO+CuO series confirm to this conclusion.

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