# Transport properties of the percolation system $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$

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Percolation behavior of normal-state resistivity and superconductivity of $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ composites have been studied. The normal-state and superconducting percolation-threshold values are found to be about 20 vol % of $YBa_2Cu_3O_{7-\delta}$ in the composite. The x-ray-diffraction and resistivity measurements on the above percolation system have shown that there is no detectable reactivity between $YBa_2Cu_3O_{7-\delta}$ and a nonmetal ceramic $YBa_2SnO_{5.5}$ even when the two substances are mixed thoroughly and sintered at 1050 °C. The implications are discussed.

## I. INTRODUCTION

The discovery of superconducting transition at elevated temperatures in ceramic materials has received tremendous attention because of its scientific and practical potential.[1-7] The study of superconducting small aggregates, clusters, and particles is very important from both a fundamental and technological standpoint. Due to the granular nature of these materials with short coherence length[8] and large penetration depth,[9] it is interesting to study the percolation and fractal properties, quantum size effects, thermal fluctuations, and size effects on superconductivity. Transport properties of normal-metal insulator percolation systems have been extensively studied over the years.[10] Recently, there were few studies on the percolation behavior of superconductor–normal-metal composites based on electrical transport and magnetization properties.[11-13] Since the resistivity ratio $\rho_s/\rho_n$ (where $\rho_s$ is the resistivity of the superconductor and $\rho_n$ the resistivity of the normal metal at room temperature) is very small for the superconductor–normal-metal composite system, the percolation model which describes the normal-state transport behavior of the composite system cannot be applied strictly to such a system. It is difficult to have a continuous network for the $YBa_2Cu_3O_{7-\delta}$ ceramic system to exist in another ceramic system without deteriorating the superconductivity of $YBa_2Cu_3O_{7-\delta}$. The $YBa_2Cu_3O_{7-\delta}$ phase requires high-temperature annealing in an oxygen atmosphere in order to retain its superconducting properties. Under such adverse conditions, most metals and insulators will react with $YBa_2Cu_3O_{7-\delta}$ and degrade the superconductivity.[12]

In this paper we report on the percolation studies on an entirely new superconductor-ceramic insulator, a $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ system based on x-ray diffraction (XRD) and electrical measurements. These studies reveal that there is no detectable reactivity between $YBa_2Cu_3O_{7-\delta}$ and a nonmetal $YBa_2SnO_{5.5}$ even when the two substances are mixed thoroughly and sintered at 1050 °C. This is a report of an ideal superconductor-insulator composite system, where the percolation model holds and at the same time superconductivity of $YBa_2Cu_3O_{7-\delta}$ is retained without deteriorating under severe heat treatment.

## II. EXPERIMENT

Pure $YBa_2Cu_3O_{7-\delta}$ has been prepared from high-purity $Y_2O_3$, $BaCO_3$, and CuO by the solid-state reaction method. The compound $YBa_2SnO_{5.5}$ has also been synthesized by the solid-state reaction method. It has a cubic structure with a lattice constant $a = 8.403$ Å. Details of preparation and characterization of the compound can be found in our previous publication.[14] The ceramic-ceramic composites are prepared by mixing different volume percentages of $YBa_2Cu_3O_{7-\delta}$ and $YBa_2SnO_{5.5}$ and sintered at temperatures in the range 950–1300 °C for 15 h. The electrical resistivity measurements were carried out by a four-probe method for the superconducting samples using a Keithley nanovoltmeter (model 181) and current source (model 220). Resistivity measurements of the composite in the higher resistance range were done by a Keithley electrometer (model 602) at room temperature. The reactivities of $YBa_2Cu_3O_{7-\delta}$ and $YBa_2SnO_{5.5}$ were studied by the x-ray-diffraction technique throughout the composition range using Ni-filtered Cu $K\alpha$ radiation.

## III. RESULTS AND DISCUSSION

The resistivities of the composites with different volume percentages of $YBa_2SnO_{5.5}$ ($V_N$) were measured in the temperature range 300–77 K (Fig. 1). For clarity and readability, the variation of $\rho/\rho_r$ with respect to temperature has been shown, where $\rho_r$ is the room-temperature resistivity of the sample. From the figure it is clear that for low $V_N$ in the composites, the resistivity shows a metallic character, but for $V_N > 70\%$, the resistivity shows a semiconductor character. The resistivity measurements indicate that the $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ composite shows superconducting transition up to 80 vol % of $YBa_2SnO_{5.5}$. Even though there is superconducting transition for 80 vol % $YBa_2SnO_{5.5}$ in the composite, its resistivity does not become zero up to a temperature of 77 K. It can be due to the absence of a superconducting network through the matrix of the composite because of the low vol % of $YBa_2Cu_3O_{7-\delta}$. Thus the superconducting percolation threshold for the $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ composite

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lies below 20 vol % of $YBa_2Cu_3O_{7-\delta}$ in the composite.

Figure 2(a) provides the normal-state resistivity of the composite as a function of the vol % of $YBa_2Cu_3O_{7-\delta}$. Figure 2(b) gives the temperature coefficient of resistivity $\alpha[=(1/\rho)(d\rho/dT)]$ at room temperature for different vol % of $YBa_2SnO_{5.5}$. Both the normal-state resistivity and temperature coefficient of resistivity showed a sharp deviation in their behavior around 80 vol % of $YBa_2SnO_{5.5}$. The percolation threshold value $V_c$ for the normal-state transport properties of the composite is around 20 vol % of $YBa_2Cu_3O_{7-\delta}$. Thus the superconducting percolation threshold and normal-state percolation threshold values of the $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ composite lie in the same range.

The normal-state transport properties of a superconductor-insulator composite can be expressed by a set of relations[15,16]

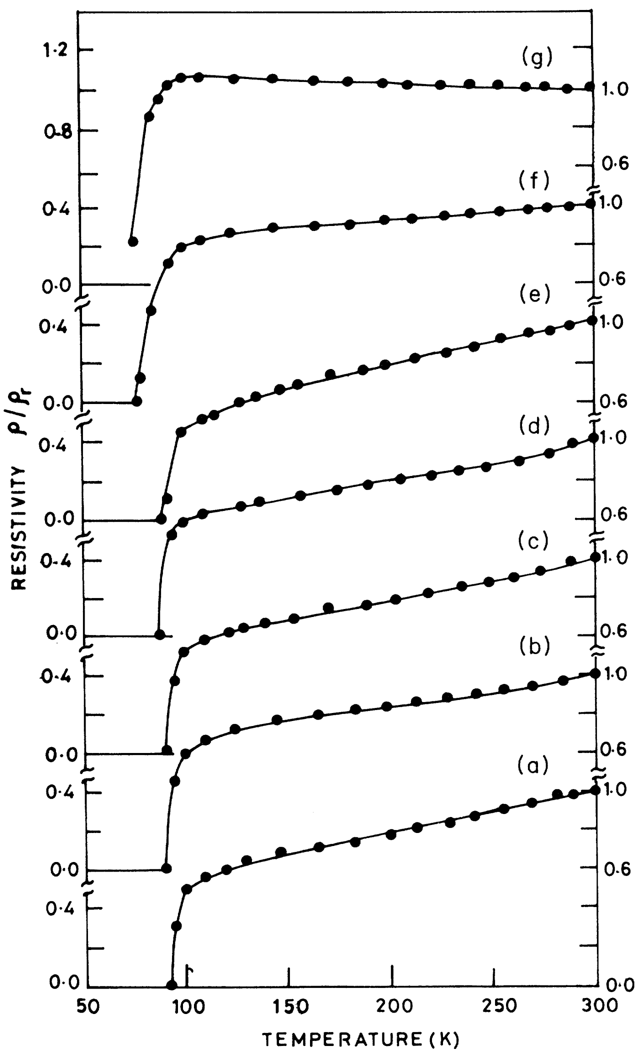


FIG. 1. Variation of normalized resistivity ($\rho/\rho_r$) with temperature for different volume percentages of $YBa_2SnO_{5.5}$ in the composite: (a) 0%, (b) 10%, (c) 20%, (d) 30%, (e) 50%, (f) 70%, (g) 80%.

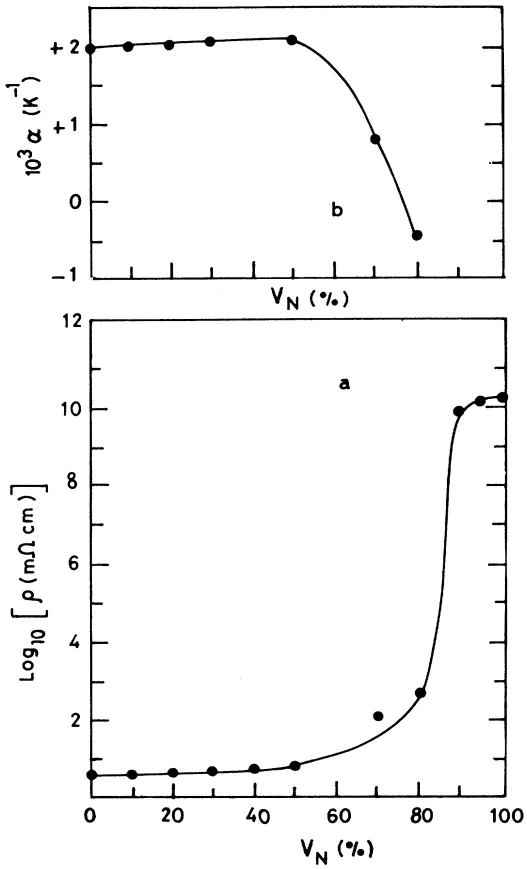


FIG. 2. (a) Variation of normal-state (room temperature) resistivity $\rho$ with different volume percentages ($V_N$) of $YBa_2SnO_{5.5}$. (b) Variation of temperature coefficient of resistivity $[\alpha=(1/\rho)(d\rho/dT)]$ at room temperature for different volume percentages ($V_N$) of $YBa_2SnO_{5.5}$.

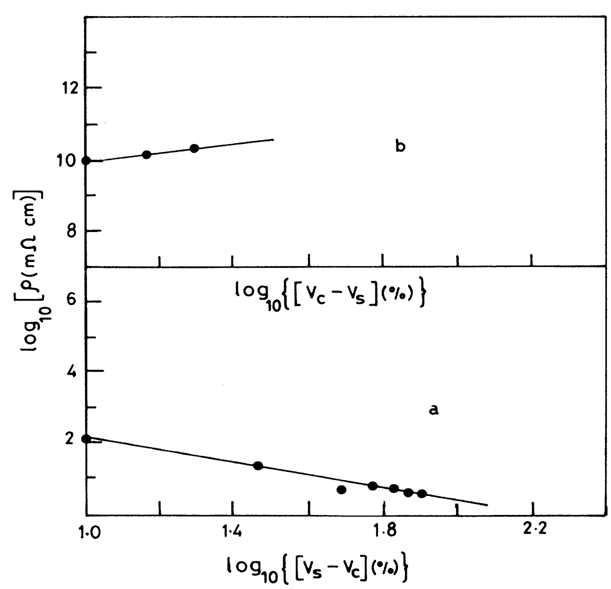


FIG. 3. (a) Log-log plot of resistivity $\rho$ with $V_s - V_c$. (b) Log-log plot of resistivity $\rho'$ with $V_c - V_s$, where $V_c$ is the percolation threshold value and $V_s$ is the volume percent of $YBa_2Cu_3O_{7-\delta}$ in the composite.

$$\rho=\rho_0(V_s-V_c)^{-t} \text{ for } V_s>V_c \ , \tag{1}$$

$$\rho'=\rho_0'(V_c-V_s)^{u} \text{ for } V_c>V_s \ , \tag{2}$$

where $\rho_0$ and $\rho_0'$ are constants, $V_c$ is the critical volume fraction at which the transport properties change drastically (called the threshold value), $V_s$ is the vol % of $YBa_2Cu_3O_{7-\delta}$ in the composite, and $t$ and $u$ are the critical exponents describing the transport properties of the composite system. The values of $\rho_0$, $\rho_0'$, $t$, and $u$ are found from the log-log plot of $\rho$ vs $V_s-V_c$ and $\rho'$ vs $V_c-V_s$. For an idealized metal-insulator composite system, the percolation threshold lies around 17% and the values of the critical exponents are around $t=1.7$ and $u=0.7$, respectively. The value of $V_c$ is taken such that the log-log plot of Eqs. (1) and (2) gives a straight line [Figs. 3(a) and 3(b)]. The values of $\rho_0$, $\rho_0'$, $t$, and $u$ are $9.77\times10^3$ m $\Omega$ cm, $5.62\times10^8$ m $\Omega$ cm, 1.8, and 1.23, respectively, from the above figure. The percolation threshold value and the superconducting percolation threshold value for the $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ composite are in the same range as expected for an idealized percolation system. Also the values of the critical exponents obtained for the above system agree reasonably well with the theoretical values.

Figure 4 shows the x-ray-powder diffraction patterns of different volume percentages of composites. From the diffraction pattern it is very clear that there is practically no reaction between the two systems, but it remains as a composite throughout the entire composition range. Figure 4(d) shows the XRD pattern for a 70 vol % of $YBa_2SnO_{5.5}$ in the composite. Although the superconducting phase in the system is very small, it gives a superconducting transition around 80 K, indicating that there exists a continuous network of superconducting paths through the matrix of the composite.

It is interesting to note that the sintering temperature of the composite increases with the increase of $YBa_2SnO_{5.5}$ volume fraction. For 70 vol % of $YBa_2SnO_{5.5}$, the sintering temperature is 1050 °C, much higher than the normal sintering temperature for $YBa_2Cu_3O_{7-\delta}$ superconductors. The studies on $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ percolation systems show that the properties of $YBa_2Cu_3O_{7-\delta}$ are not degraded, and as seen by x-ray studies there is no detectable reaction for $YBa_2Cu_3O_{7-\delta}$ even at an elevated temperature of 1050 °C. Due to the nonreactive nature of $YBa_2Cu_3O_{7-\delta}$ with $YBa_2SnO_{5.5}$ at elevated temperatures, it may be possible to deposit thin films of $YBa_2Cu_3O_{7-\delta}$ on $YBa_2SnO_{5.5}$ substrates. Another important aspect of the investigation is the fabrication of artificial superlattice structures made of $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ structures with varying modulation lengths. Successful fabrication of such superlattice structures paves the way to understand the fundamental mechanism for high-temperature superconductivity through proximity-effect studies.[17]

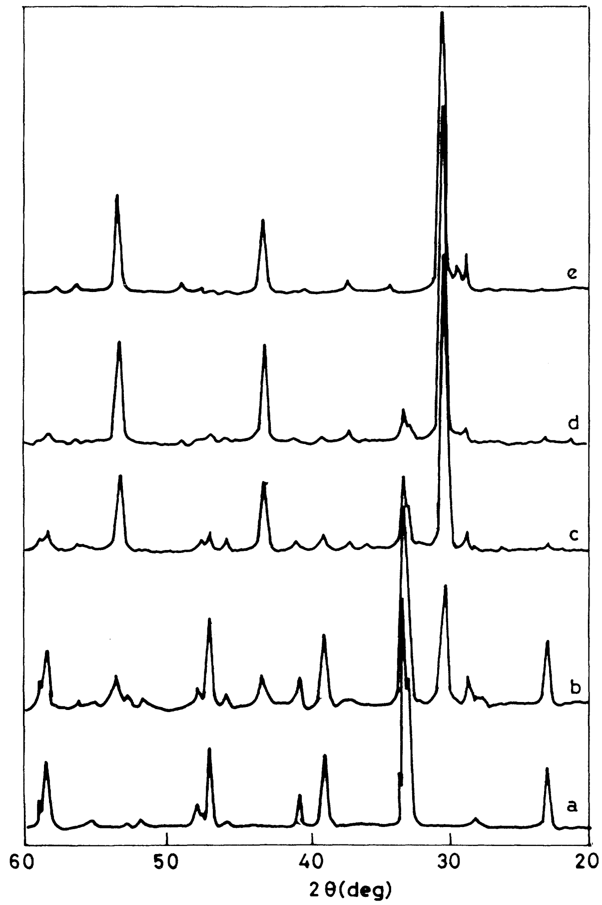


FIG. 4. X-ray powder diffraction pattern for different volume percentages of $YBa_2SnO_{5.5}$ in the composite. (a) 0%, (b) 30%, (c) 50%, (d) 70%, (e) 100%.

## IV. CONCLUSION

The normal-state percolation and superconducting percolation threshold values in $YBa_2Cu_3O_{7-\delta}$-$YBa_2SnO_{5.5}$ are found to be the same, that is, about 20 vol % of $YBa_2Cu_3O_{7-\delta}$. The critical exponents describing the transport behavior of the composite system match fairly well with those for a perfect metal-insulator composite system. These studies have shown that there is no detectable reactivity between $YBa_2Cu_3O_{7-\delta}$ and a nonmetal ceramic $YBa_2SnO_{5.5}$ even under severe heat treatment, making $YBa_2SnO_{5.5}$ a possible substrate material for $YBa_2Cu_3O_{7-\delta}$ films. However, this aspect needs further detailed investigation.

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