FLUX PINNING BY NANO PARTICLES EMBEDDED IN POLYCRYSTALLINE Y-123 SUPERCONDUCTORS

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ABSTRACT
Bulk superconductor samples of YBa$_2$Cu$_{3-x}$Zn$_x$O$_{7-\delta}$ with $x = 0, 0.01, 0.03$ are synthesized by solid-state reaction route. The structural characterisation of all samples has been carried out by x-ray-diffraction (XRD) and transmission electron microscopy (TEM) techniques. The x-ray diffraction patterns indicate that the gross structure/phase of YBa$_2$Cu$_{3-x}$Zn$_x$O$_{7-\delta}$ do not change with the substitution of Zn up to $x=0.03$. In TEM investigations of Zn-doped Y-based cuprates a number of ZnO nano-flower and nano-rod of Y-211 phase are found dispersed in regular YBa$_2$Cu$_3$Zn$_x$O$_7$ matrix. These dispersed nano-flowers of ZnO and nano-rods of Y-211 phase may serve as flux-pinning centers. These pinning centers enhance critical current density ($J_c$) value of these HTSC samples.

Keywords: High Temperature Superconductor; Zn-Doped YBCO; Solid State Reaction; Electron Microscopy; Flux Pinning; Critical Current Density ($J_c$).

1. INTRODUCTION
High critical temperature (high-Tc) superconductors have been the subject of extensive studies carried out to improve their superconducting properties. After the critical transition superconducting temperature (Tc), the critical current density ($J_c$) is the most important parameter for potential applications of high-Tc superconductors at liquid-nitrogen temperature under an applied magnetic field. The improvement of $J_c$ and its behavior under a magnetic field $J_c(H)$ can be obtained by introducing efficient pinning centers with a size (matching the coherence length) that can suppress the flux flow.

Various methods have been used to introduce effective artificial pinning centers and to increase their number [1-31]. Flux pinning by non-superconducting inclusions and crystal defects is well-documented for YBa$_2$Cu$_{3}$O$_{7-\delta}$ (Y-123 or YBCO for brevity) [1-4]. Many works have indicated that twin boundaries [5, 6], dislocations and stacking faults [1, 7] are possible pinning centers.

Precipitates dispersed into the superconducting matrix are often found to be an effective pinning center. Such precipitates can be obtained by precipitation from a single phase superconductor or by introduction of secondary phase [8-19].

The embedding of nanometre-sized particles [20-26] into the superconducting matrix as flux pinning centres was the goal for a long time since the coherence length of the high-Tc superconductors (Y-123) is so small (20nm to 300nm).

Nano-sized non-superconducting regions can also be introduced into YBCO bulk superconductors by chemical substitution of atoms in the Y123 lattice (mainly the Cu atoms in the CuO chains [27] or the CuO planes [28-30]). These artificially created pinning centres can be effective for enhanced flux pinning in YBCO at intermediate magnetic fields and high temperatures.

The central aim of the present work is to investigate the structural and microstructural changes due to substitution of Zn at Cu site of Y-123 and their possible correlation with superconducting properties (such as critical current density).

2. EXPERIMENTAL DETAILS
Samples with nominal composition YBa$_2$Cu$_3$, $x$Zn$_x$O$_{7-\delta}$ (where $x = 0, 0.01, 0.03$) were synthesised by standard solid state reaction method. The appropriate ratio of the constituent oxides or carbonate i.e. Y$_2$O$_3$ (99.9%, Alfa Aesar), CuO (99.99%, Alfa Aesar), ZnO (99.0%, Alfa Aesar) and BaCO$_3$ (99.0%, Alfa Aesar) were thoroughly mixed and ground for several hours (2 to 4 hrs) with the help of mortar and pestle. After regrinding and mixing, the powder was kept in a platinum or alumina crucible and heated (calcined) at 875°C. The calcinations step served to decompose the carbonate and starting
material to interdiffuse for phase formation and chemical homogeneity. After calcinations the material was again ground to subdivide any aggregated products and to further enhance chemical homogeneity. These steps were repeated 3 to 4 times for better homogeneity and phase purity.

The homogeneous powder thus formed was converted into form of pellets before sintering. For this we employed the most widely used technique i.e. dry pressing, which consists of filling a die with powder and pressing at 400 Kg/cm² into a compacted disc shape. In this way several pellets with varying thickness (1mm to 3mm) were prepared. Finally these pellets were put into alumina crucibles and sintered at about 920±5°C in air. The heating rate to the sintering temperature was about 100°C/hour. After sintering at 920±5°C final annealing was carried out in oxygen atmosphere at partial pressure 10⁻¹ atm, at temperature 570°C for 14 hrs in order to maximize the incorporation of oxygen. The annealing temperature is compromise between higher temperatures where oxygen diffusion rates are higher and lower temperatures where the thermodynamic equilibrium favours higher oxygen contents.

Some important factors that can determine ‘sample quality’ during sample preparation are the stoichiometry, the mixing procedure, the sintering temperature, the oxygen flow during annealing and the rate of sample cooling. If the sample preparation is done with care, a better understanding of the roles these major factors play should ensure that final samples are of high quality.

The structure and phase purity of the powder sample ground from sintered pellets were examined by x-ray diffraction technique using a powder x-ray diffractometer (18 KW, Rigaku Japan) with CuKα radiation, λ = 1.5408Å. The diffraction data were collected over the diffraction angle range of 2θ= 0-90° by step scanning with a scanning rate 2°/minute. The structural/ microstructural characteristics was explored by transmission electron microscope (TEM, Tecnai 20 2G) in both the imaging and diffraction modes.

Finally in order to see the effect of Zn doping on the physical properties, the transport critical current density (Jc) value has been measured by standard four-probe method as potential difference of 1µV/cm appears across the sample by increasing current at temperatures ranging from 77 to 60 K.

3. RESULTS & DISCUSSIONS

Fig. 1 shows the powder x-ray diffraction patterns of YBa₂Cu₃₋ₓZnₓO₇₋₅ with x = 0, 0.01 and 0.03. The powder diffraction result of these samples showing major phase of Y-123. All the peaks were indexed on the basis of Y-123 structure. The crystal structure was found to be orthorhombic (Pnmm space group) with Zn substitution up to x=0.03.

The Zero-resistance critical temperature Tc (R=0) decreases systematically with the increase in concentration of zinc. The values of Tc (R=0) for the samples with zinc concentrations x= 0, 0.01, and 0.03 are 91K, 60K and 55K respectively.

In order to explore the effect of Zn doping on the microstructural characteristics in YBa₂Cu₃₋ₓZnₓO₇₋₅ superconductors, transmission electron microscopy was employed in imaging and diffraction modes.

In TEM investigation of Zn-doped Y-based cuprates a number of ZnO nano-flower are found dispersed in regular YBa₂Cu₃₋ₓZnₓO₇₋₅ matrix as shown in Figure 2. These dispersed nano structure may act as flux pinning centers that result high transport (inter-grains) critical current density Jc ~ 10³ A / cm² at 50K.
As the most prominent pinning center in melt textured high-T\textsubscript{c} superconductors is the insulating Y\textsubscript{2}BaCuO\textsubscript{5} (Y-211) particle being created during the growth of YBCO and providing flux pinning. Figure 3(a) shows, a nano-rod of this Y-211 phase and its selected area diffraction (SAD) pattern is shown in Figure 3(b).

The variation of the transport critical current density \( (J_c) \) of YBa\textsubscript{2}Cu\textsubscript{3-x}Zn\textsubscript{x}O\textsubscript{7-δ} with different Zinc concentration revealed one order enhancement in \( J_c \) value as shown in fig. 4. In Zinc free sample critical current density \( (J_c) \) has been measured to be about \( 1.5 \times 10^2 \) A/cm\textsuperscript{2} but \( J_c=2.57 \times 10^3 \) A/cm\textsuperscript{2} has been found for zinc doped \( (x=0.03) \) sample.

A serious handicap of the superconducting cuprates is the easy motion of their vortices, so that their critical current density tends to decrease dramatically when the temperature approaches \( T_c \) or when a high magnetic field is applied. People speculated that one could improve the critical current in crystals by introducing defects. A magnetic field penetrates a type-II superconductor in the form of vortices. Each vortex, carrying a flux quantum \( \Phi=h/2e \), consists of a cylindrical core of radius \( \xi \), the coherence length of the material, and a current circulating around the core out to a distance \( \lambda \), the material penetration depth. When a current flows in the superconductor, the Lorentz force produces energy dissipation and consequently the disappearance of superconductivity.

The mobility of the vortices can be minimized by introducing efficient pinning centers that pin the vortices. Thus the critical current density \( J_c \) depends on flux pinning of vortices. Nano-defects will be the best pinning centers since high Tc superconductor of type Y-123, Tl-2212 and Tl-2223 have a coherent length of range 20nm to 300nm. In present Zn doped Y-based system some nano-flowers of ZnO and nano-rods of Y-211 phase have already been observed in electron microscopic investigations. These nano-flowers and nano-rods may act as effective flux pinning centers. These pinning centers enhance
critical current density (Jc) value of these HTSC materials as observed in our transport Jc measurements.

4. CONCLUSION

We have successfully prepared samples of type YBa$_{2}$Cu$_{3-x}$Zn$_{x}$O$_{7-δ}$ with x = 0, 0.01 and 0.03 by standard ceramic method. The partial replacement of Cu by Zn does not affect the orthorhombic structure of the Y-123 phase. The structural/microstructural characteristics were explored through transmission electron microscopy (TEM). An interesting result has been found in TEM study that the dispersion of nano-flowers and nano-rod of ZnO and Y-211 phase respectively in local region of Zn-doped Y-based cuprates. These dispersed nano-flowers of ZnO and nano-rods of Y-211 phase possibly behave as flux pinning centers. The critical current density Jc increases as Zinc content increases as a result of the flux pinning centers of nano-flowers of ZnO and nano-rods of Y-211 phase.

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