Effect of Co_{0.5}Zn_{0.5}Fe₂O₄ Nanoparticle on AC Susceptibility and Electrical Properties of YBa₂Cu₃O₇₋₈ Superconductor

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Magnetism and superconductivity are mutually exclusive phenomena and their interaction is an interesting topic to study. In this work the effect of $Co_{0.5}Zn_{0.5}Fe_2O_4$ (CZFO) nanoparticle with size 20-50 nm on YBa₂Cu₃O_{7- δ}(YBCO) is reported. The samples were prepared using solid state reaction with starting composition YBa₂Cu₃O_{7- δ}(Co_{0.5}Zn_{0.5}Fe₂O₄)_x with x = 0 to 0.4 weight percent (wt. %). All samples exhibited single YBCO phase as shown from the XRD patterns. The grain size was reduced with CZFO addition. The transition temperature (T_c) from resistance measurements showed a slight increase from 90 K (x = 0) to 91 K (x = 0.1) followed by a suppression for $x \ge 0.2$. The peak temperature, T_p of the imaginary part of the susceptibility χ ", was around 79 to 76 K in the x = 0 to 0.3 samples. However, a drastic decrease to 60 K was observed in the x = 0.4 sample indicating weakening of intergrain coupling which resulted in coupling losses as CZFO was added. The x = 0.1 sample showed the highest T_c and the critical current density, J_c among all the samples studied. These results were compared with other materials addition to YBa₂Cu₃O_{7- δ}.

Keywords: coherence length; penetration depth; nanoparticles; intergrain current density

1. INTRODUCTION

The exclusive nature of magnetism and superconductivity make them interesting topic to study. The interaction between flux line network and magnetic texture can improve the critical current density, J_c through the addition of magnetic nanomaterial [1, 2]. Addition of antiferromagnetic γ -Fe₂O₃,

ferromagnetic Fe₃O₄ and diamagnetic ZnO in Bi-Sr-Ca-Cu-O enhanced the critical current density [3-5]. Addition of nanosized PbO in YBa₂Cu₃O_{7- δ} (YBCO) showed enhancement of current density and transition temperature [6]. Nanosized particle addition in high- T_c superconductors can improve J_c and the transition temperature, T_c [7-11]. However, ferromagnetic nanoparticles such as Co₃O₄ [12] and CoFe₂O₄ [13] suppressed the transition temperature of YBCO.

Complex magnetic oxides show a wide range of properties arising from the strong interplay among the physical properties making them ideal for flux pinning centers [8, 14]. Cu_{0.5}Tl_{0.5}Ba₂Ca₂Cu₃O₉, added with 0.08 weight percent (wt. %) Co_{0.5}Zn_{0.5}Fe₂O₄ (CZFO) showed improvement in transition temperature by 3.35 % and an increase in hole concentration [15]. CZFO is superparamagnetic in nanosize and when prepared with a template it is ferromagnetic [16].

 J_c was suggested to increase when the magnetic nanoparticles size, *d* is larger than the coherence length ξ but smaller than the penetration depth, λ [9]. The flux pinning is suggested to increase when the size of the nanoparticles approaches ξ [17]. The coherence length ξ for YBCO is about 1.65 nm and the penetration depth λ is about 156 nm [18].

In this paper, Co_{0.5}Zn_{0.5}Fe₂O₄ nanoparticles with size d = 20 - 50 nm were added into YBa₂Cu₃O_{7- δ . This size *d* was chosen because it is between the coherence length and penetration depth, $\xi < d < \lambda$ of the YBCO [8]. In this research we investigated the effect of this nanoparticle on the properties of YBCO superconductor. The electrical resistance was measured along with the structural properties. The AC susceptibility $\chi = \chi' + \chi''$ behavior and the inter-granular critical current density at the peak temperature, T_p of the complex susceptibility χ'' , $J_c(T_p)$ is also reported in this paper [8]. The results were compared with previous reports on nanoparticle addition in YBCO.}

2. EXPERIMENTAL DETAILS

The material was prepared using high purity (> 99.9 %) Y₂O₃, BaCO₃ and CuO with starting chemical formula YBa₂Cu₃O_{7- δ}. The powders were heated at 900 °C for over 24 h with intermittent grindings. Nanosized Co_{0.5}Zn_{0.5}Fe₂O₄ (from Inframat Advanced Materials) was then added to the resultant powders with starting formula YBa₂Cu₃O_{7- δ}(Co_{0.5}Zn_{0.5}Fe₂O₄)_{*x*} where *x* = 0, 0.1, 0.2, 0.3 and 0.4 wt. %. The powders were pressed into pellets of about 12 mm diameter and 2 mm thickness and heated at 900 °C for 24 h.

The resultant phases were identified by using the X-ray diffraction method with Bruker D8 Advanced diffractometer. Lattice parameters were calculate using at least 10 diffraction peaks. The field emission scanning electron micrographs (FESEM) were recorded using a Merlin Gemini scanning electron microscope [8]. A Philips transmission electron microscope (TEM) model CM12 was used to record the average size of the $Co_{0.5}Zn_{0.5}Fe_2O_4$ starting nanoparticles.

The four-probe method was used to measure the electrical resistance. Silver paint was used as electrical contacts. A closed cycle refrigerator from CTI Cryogenics (Model 22) and temperature controller from Lake Shore (Model 330) were used for low temperature measurements. A Cryo Industry AC susceptometer model number REF-1808-ACS was used for the AC susceptibility measurements. The frequency used was 295 Hz and magnetic field was 400 A/m. Bar-shaped sample with cross section

dimension of approximately 2 mm × 2 mm was used. The critical current density at the peak temperature T_p of χ " was calculated with formula $J_c(T_p) = H/(lw)^{1/2}$, where *H* is the applied field, *l* and *w* are the cross-section dimensions of the sample [19].

3. RESULTS AND DISCUSSION

All samples showed single-phase YBa₂Cu₃O_{7- δ} (space group Pmmm). Figure 1(a) shows the XRD patterns for x = 0, 0.1 and 0.2. Figure 1(b) shows the patterns for x = 0.3 and 0.4. No systematic change in the lattice parameters was observed (Table 1). The lattice parameters are a = 3.826 Å, b = 3.893 Å and c = 11.702) Å for x = 0 sample. The internal lattice strain is determined by the ratio of the lattice parameter c/a and c/b. All samples showed $c/a \sim 3.06$ and $c/b \sim 3$. This shows that no change in the internal lattice strain in these samples. TEM micrographs of CZFO shows that the particle size was between 20 and 50 nm (Figure 2). The SEM micrographs for x = 0, 0.1 and 0.4 show that the grain size increased as CZFO was added i.e. 3, 5, and 10 µm, respectively (Figure 3).



Figure 1. XRD patterns of CZFO added YBa₂Cu₃O_{7- δ} samples (a) x = 0, 0.1 and 0.2 and (b) x = 0.3 and 0.4



Figure 2. TEM micrograph of CZFO



Figure 3. SEM micrograph of (a) x = 0, (b) x = 0.1 and (c) 0.4 wt. % samples

The resistance versus temperature curves showed that T_c was somewhat affected by CZFO. The onset transition temperature $T_{c-onset}$ increased for x = 0.1 sample and then decreased with further addition (Figure 4). The x = 0.2 and 0.3 samples showed slight decrease in T_c . However, the x = 0.4 sample showed a drastic drop in $T_{c-onset}$ (73 K) and T_{c-zero} (63 K). The transition width, ΔT_c is about 5 K for x = 0, 0, 1, 0.2 and 0.3 and 10 K for x = 0.4. The large variation in the T_c of individual superconducting grain as CZFO was increased in x = 0.4 may cause the increase in ΔT_c .



Figure 4. Electrical resistance versus temperature curves of YBa₂Cu₃O_{7-δ} samples added with CZFO nanoparticles

AC susceptibility data showed that the transition temperature $(T_c\chi')$ decreased slightly with the addition of CZFO (Figure 5). The non-added sample showed $T_c\chi' = 92$ K and other samples showed transition between 90 and 91 K. The sudden drop in the real part χ' of the complex susceptibility $(\chi = \chi' + \chi'')$ below $T_c\chi'$ is due to diamagnetic shielding. The peak in χ'' indicates AC losses [8].

Two loss peaks (Figure 5) that include a broad peak at low temperature (T_p) are due to coupling losses and at higher temperature the narrow peak indicates intrinsic losses. All of our samples showed the higher temperature peak which coincide with the sharp drop in the real part of the susceptibility χ' . This showed a large inter-granular potential and flux penetration into the grains of the sample [20]. T_p decreased as CZFO was added due to lower intergrain coupling of the superconducting phase. T_p of the x = 0.4 sample shifted to lower temperature (60 K) which may be due to the weak pinning force strength. The weaker the pinning, the larger is the shift in T_p . At T_p , the AC field amplitude is equal to the full flux penetration field [8, 20].



Figure 5. The AC susceptibility $\chi = \chi' + \chi''$ versus temperature graph of YBa₂Cu₃O_{7- δ} samples added with CZFO nanoparticles (a) x = 0, 0.1 and 0.2 and (b) x = 0.3 and 0.4. Inset shows χ' near the transition temperature.

If the applied field *H* is equal to the first full penetration field, the flux line will penetrate the superconductor fully and the losses are maximal [19]. The critical current density as a function of temperature can be determined from this fact. i.e. using the critical state model. The transport critical current density at the peak temperature $J_c(T_p) = 16 - 18 \text{ A cm}^{-2}$. The x = 0.1 sample showed the highest $J_c(76 \text{ K}) = 18 \text{ A cm}^{-2}$. This work showed that this sample exhibited the best result in terms of transition temperature and critical current density. Table 1 shows T_{c-zero} , $T_{c-onset}$, AC susceptibility transition temperature $T_{c\chi'}$, peak temperature of χ'' , T_p , and the intergrain critical current density at T_p , $J_c(T_p)$.

<i>x /</i> wt. %	<i>a</i> / Å	<i>b</i> / Å	<i>c</i> / Å	T _{c-onset} / K	T _{c-zero} / K	<i>T</i> _c χ' / K	<i>T</i> _p / K	$J_{\rm c}(T_{\rm p}) / A{\rm cm}^{-2}$
0.00	3.826	3.893	11.707	90	85	92	79	16
0.10	3.830	3.893	11.716	91	86	91	76	18
0.20	3.828	3.896	11.693	86	78	90	77	17
0.30	3.827	3.895	11.716	89	84	90	77	16
0.40	3.834	3.895	11.713	73	63	90	60	16

Table 1. Lattice parameters, $T_{c\text{-zero}}$, $T_{c\text{-onset}}$, $T_{c\chi}$, T_{p} , $J_{c}(T_{p})$ for YBa₂Cu₃O_{7- δ} samples added with CZFO nanoparticles

The effect of various addition of nanoparticles on YBCO is shown in Table 2. Superparamagnetic CZFO showed slight improvement in J_c and T_{c-zero} . Antiferromagnetic material such as FeF₂ [21], ferrimagnetic such as Cr₂S₃ [22] and CdTe [23] which is diamagnetic [24, 25], enhanced T_c of YBa₂Cu₃O_{7- δ} while ferromagnetic materials such as Co₃O₄ [12] and CoFe₂O₄ [13] suppressed the transition temperature.

Addition	<i>x</i> / wt.%	$T_{\text{c-onset}} / \mathrm{K}$	Ref.
Non-Added YBa ₂ Cu ₃ O _{7-d}	0	90	This work
$Co_{0.5}Zn_{0.5}Fe_2O_4$ (superparamagnetic)	0.1	91	This work
Cr ₂ S ₃ (ferrimagnetic)	0.05	93	[23]
Co ₃ O ₄ (ferromagnetic)	0	92	[12]
FeF ₂ (antiferromagnetic)	0.03	92	[23]
CdTe (diamagnetic)	0.07	93	[24]
CoFe ₂ O ₄ (ferromagnetic)	0	92	[13]

Table 2. The effect of various addition of nanoparticles on YBCO

In conclusion, the effects of CZFO on YBa₂Cu₃O_{7- δ} have been investigated. CZFO enhanced the transition temperature and critical current density for low addition level (*x* = 0.1). The peak temperature of χ " (*T*_p) and *J*_c(*T*_p) decreased as CZFO was added indicating weakened flux pinning and intergrain coupling. By comparing with previous works, in general ferromagnetic materials suppressed *T*_c while other materials such as superparamagnetic, ferrimagnetic, antiferromagnetic and diamagnetic particles enhanced *T*_c and *J*_c for low-level addition.

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