SYNTHESIS AND CHARACTERIZATIONS OF $YBa_2Cu_3O_{7-\delta}$ SUPERCONDUCTOR WITH ADDED Al_2O_3 NANOPARTICLES VIA CITRATE-NITRATE AUTO-COMBUSTION REACTION

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ABSTRACT

Superconductor materials are renowned to conduct electricity at zero resistance and capable to expel magnetic flux. It can be used in developing efficient wire cables, magnetic energy storage and levitation technologies. Thus, the high temperature superconductor $YBa_2Cu_3O_{7-\delta}$ with added Al_2O_3 nanoparticles was synthesized via citrate-nitrate auto-combustion reaction process. The novelty of this research work is the citrate-nitrate auto-combustion reaction method consumed less energy and time compared with other conventional synthesis methods for processing of composite superconductor oxides and produced well distribution of Al_2O_3 nanoparticles in $YBa_2Cu_3O_{7-\delta}$ superconductor. The auto-combustion reaction transformed the formulated precursor citrate-nitrate gel into very fine ashes. It yielded Al_2O_3 and $YBa_2Cu_3O_{7-\delta}$ phases after calcination process which was further heat treated to achieve superconductivity. The reactions during synthesis processes were investigated through the thermal evaluations. The effects of different concentration of Al_2O_3 nanoparticles on the structure, superconducting, magnetic and mechanical properties of $YBa_2Cu_3O_{7-\delta}$ were investigated and appraised. The sustained orthorhombic structure in each sample contributed to consistency in superconducting transition temperature while the flux pinning forces provided by the non-superconducting nanoparticles improved the critical current density. Furthermore, the mechanical hardness of the samples was also influenced by the addition of nanoparticles. This work shows that the citrate-nitrate auto-combustion reaction is an effective method to introduce Al_2O_3 as nanoparticles homogeneously distributed in the $YBa_2Cu_3O_{7-\delta}$ superconductor.

SINTESIS DAN PENCIRIAN SUPERKONDUKTOR $YBa_2Cu_3O_{7-\delta}$ DITAMBAH ZARAH NANO Al_2O_3 MELALUI TINDAK BALAS PEMBAKARAN - AUTOMATIK SITRAT-NITRAT.

ABSTRAK

Bahan-bahan superkonduktor telah diketahui dapat mengalirkan arus elektrik pada rintangan sifar dan menyingkir fluks magnet. Ia boleh digunakan dalam membangunkan teknologi wayar kabel efisien, penyimpanan tenaga dan pengapungan magnet. Oleh itu, superkonduktor suhu tinggi $YBa_2Cu_3O_{7-\delta}$ terkandung zarah nano Al_2O_3 telah disintesis melalui tindak balas pembakaran-automatik sitrat-nitrat. Keaslian kerja penyelidikan ini ialah kaedah sintesis ini lebih menjimatkan tenaga dan masa berbanding kaedah lama dalam penghasilan komposit superkonduktor oksida dan dapat menghasilkan serakan seragam zarah nano Al_2O_3 dalam superkonduktor $YBa_2Cu_3O_{7-\delta}$. Pembakaran-automatik telah menukarkan gel sitrat-nitrat yang telah diformulasi kepada abu-abu yang sangat halus. Ianya menjadi fasa-fasa Al_2O_3 dan $YBa_2Cu_3O_{7-\delta}$ setelah dikalsin sebelum dirawathaba bagi mencapai kesuperkonduksian. Setiap tindak balas yang berlaku semasa proses sintesis ini diperincikan melalui kaedah penilaian terma. Segala kesan akibat penggunaan kepekatan berbeza zarah nano Al_2O_3 terhadap struktur, sifat kesuperkonduksian, kemagnetan dan sifat mekanikal $YBa_2Cu_3O_{7-\delta}$ dikaji dan ditaksirkan. Struktur ortorombik setiap sampel menyumbang kepada suhu peralihan kesuperkonduksian yang konsisten manakala daya pengepin yang diperolehi daripada zarah nano bukan superkonduktor telah menambahbaik ketumpatan arus genting. Selain itu, kekerasan mekanikal sampel-sampel juga telah terpengaruh dengan penambahan zarah nano ini. Kajian ini menunjukkan bahawa kaedah pembakaran-automatik sitrat-nitrat merupakan kaedah berkesan untuk menghasilkan Al_2O_3 sebagai bahan zarah nano tertabur secara seragam dalam superkonduktor $YBa_2Cu_3O_{7-\delta}$.

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Refined XRD patterns (x_{mol} = 0.08 and 0.10)

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LIST OF SYMBOLS AND ABBREVIATIONS

Symbol	Description
a	lattice constant at x direction
A	cross section of the sample
b	lattice constant at y direction
B_a	applied magnetic field
B_C	critical magnetic field
B_i	interior magnetic field
B_r	remnant flux density
c	lattice constant at z direction
c/n	citrate to nitrate ratio
SSP	solid-state processing
Cp	heat capacity
CP	co-precipitation
d	grain size
D	thickness of slab
δ	oxygen deficiency
DTA	differential thermal analysis
E	electric field
EDX	energy dispersive X-ray
E_F	fermi energy
emf	electromotive force
FESEM	field emission scanning electron microscope
F_L	Lorentz force

F_n	free energy at normal state
F_p	pinning force
f_p	basic pinning force
F_s	free energy at superconducting state
H (P)	heat required by products
H (R)	heat required by reactants
B_C, H_C	critical magnetic field
B_{C2}, H_{C2}	upper critical applied magnetic field
H_V	Vickers hardness
J_C	critical current density
J_s	density of superconducting current
k_B	Boltzmann's constant
l	length of superconductor slab
L	Phase transformation entalphy
m	mass of electron
MO	metal oxide
MOH	metal hydroxide
MPMS	magnetic properties measurement system
n_i	stoichiometry ratio of reactants coefficient
n_j	stoichiometry ratio of products coefficient
n_n	number of normal electron
n_o	average number of electrons
N_p	pinning center density
n_s	number of superconducting electron
P_j	products
Φ_o	flux quantum

R	resistance
R_i	reactants
RT	room temperature
SQUIDs	superconducting Quantum Interference Devices
t	width of superconductor slab
T_0	initial temperature
T_{ad}	adiabatic temperature
T_C	superconductor critical temperature
T_{com}	combustion temperature
TGA	thermogravimetric Analysis
T_{ig}	ignition temperature
U_o	flux creep potential
U_p	flux pinning potential
V_f	volume fraction of pinning center
v_s	velocity of superconducting electron
x_{mol}	concentration of Al_2O_3 nanoparticles in the sample
XRD	X-ray diffraction
YSZ	yttrium stabilized zirconia
ΔE	energy gap of Cooper pair fluid
ΔH	heat needed for ignition
ΔM	magnetization gap
ΔQ	heat loss
λ	penetration depth
μ_r	relative permeability

ξ	superconductors coherence length
ξ_{ab}	coherence length parallel to *a*-*b* plane
ξ_c	coherence length parallel to *c* plane
ξ_o	intrinsic coherence length
ρ	resistivity
σ	conductivity
τ	crystallite size
ψ	superconducting order parameter
κ	Ginzburg-Landau parameter

CHAPTER 1

INTRODUCTION

1.1 Research background

Superconductivity is a promising technology to prevent energy losses attributed from electrical resistivity. This technology attracts much attention to the researches due to two main reasons: the electricity can be conducted at zero resistivity and the magnetic flux can be totally repelled out from the body of superconducting material. Since discovered in 1911, superconductivity was only been found in elements and alloys where the critical temperature (T_C) of these materials are very low (Onnes, 1991). In 1986, the lanthanum cuprate was found to be the first compound material to exhibit superconductivity (Muller and Bednorz, 1986). This breakthrough was followed by discovery of yttrium barium copper oxide ($YBa_2Cu_3O_{7-\delta}$) superconducting compound in 1987 which brought a great excitement within the scientific community because this material can conduct electricity without having resistivity at temperatures above 77 K. This is the temperature at which nitrogen liquefies, thus $YBa_2Cu_3O_{7-\delta}$ changed the perspective of the applications of superconductivity and opened up the possibility for numerous advancements of technologies (Wu et al., 1987).

Categorized as type II superconductor, $YBa_2Cu_3O_{7-\delta}$ shows perfect diamagnetism at below lower critical fields (H_{C1}), allows penetration of quantized magnetic flux (vortex) at higher fields, and loss superconductivity at above higher critical fields (H_{C2}). It means that the coherence of superconducting state is preserved even in the presence of weak currents and magnetic fields below H_{C2}, and makes it practical to be used in stronger magnetic fields compared with conventional superconductors.

1.2 Problems statement

Theoretically, $YBa_2Cu_3O_{7-\delta}$ has excellent ability to carry higher critical current density (J_C) due to having very high H_{C2} compared with H_{C1}. However, when electrical current is flowed in applied magnetic field of $H_{C1} < H < H_{C2}$, the interaction between vortex and current flow results in the motion of vortex due to Lorentz force which can cause energy loss. At this point, only a small density of current limits by J_C is permitted to flow through this superconductor before losing energy. Generally the J_C can be increased if the vortex is prevented from moving. This can be achieved by pinning them with suitable non-superconducting point as known as pinning centre materials. Pinning centre materials in $YBa_2Cu_3O_{7-\delta}$ can be created either by inducing defects through irradiation techniques and chemical doping or by introducing second phase particles having nanometer size. In chemical doping, elements such as calcium (Ca), potassium (K), silver (Ag) and aluminium (Al) are diffused into $YBa_2Cu_3O_{7-\delta}$ structure (Giri et al., 2005, Celebi et al., 2000, Sen et al., 1990 and Zhang et al., 1995). These elements locally modify the crystallinity of the structure and generate defects such as twins, tweed, and inhomogeneous micro-defects to pin the vortices. The J_C can be improved by chemical doping but in return this method may reduce the T_C, since the orthorhombicity of $YBa_2Cu_3O_{7-\delta}$ is being altered; hence, decreased.

Thus for some reasons, introduction of nanoparticles in $YBa_2Cu_3O_{7-\delta}$ has generated a great interest among researchers. This method represents an easy controlled, non-destructive and efficient tool for improving the mechanical, structural and superconducting properties of $YBa_2Cu_3O_{7-\delta}$ compounds. Various nanometer particles; such as, silicon carbide (SiC), zirconia (ZrO_2), yttrium oxide (Y_2O_3), cerium dioxide (CeO_2), tin oxide (SnO_2) and aluminum oxide or Alumina (Al_2O_3) has been reported to add into $YBa_2Cu_3O_{7-\delta}$. All of these nanoparticles were acted as additional pinning centre and resulted in an increase of the J_C in higher magnetic fields (Guo et al., 1999, Zhang and Evetts, 1993, Goswami et al., 2007, Lee et al., 2001, He et al., 2001 and Mellekh et al., 2006). In order to effectively pin the vortices, nanoparticles should possess features including high density or uniform distribution. Nanoparticles having size of 3-10 nm is equal to the coherence length (ξ) of $YBa_2Cu_3O_{7-\delta}$ and able to be located at CuO_2 planes (Moutalibi and M'chirgui, 2007).

Among the added nanoparticles, Al_2O_3 is more attractive to be selected as the pinning centre material in $YBa_2Cu_3O_{7-\delta}$. Beside the capability to pin the vortex motions, Al_2O_3 nanoparticles are also selected due to its availability in nanometer size, easy fabrication route, higher thermal stability, higher density, better hardness and lower cost. In 1987, the effects of Al substitution for yttrium (Y) or copper (Cu) sites in $YBa_2Cu_3O_{7-\delta}$ were accidently found. For long period of high temperature calcination process, the use of alumina crucibles results in the incorporation of Al in crystals having composition of $YBa_2Cu_{3-x}Al_xO_y$. X-ray diffraction (XRD) analysis reveals that such substitution does not lead to changes in the structural symmetry of $YBa_2Cu_3O_{7-\delta}$ but the orthorhombicity of the system is decreased with increasing Al content (Zhang et al., 2005). The value of T_C decreases with increasing Al. This finding was confirmed by Zhang and his colleagues as they purposely added Al_2O_3 powder into $YBa_2Cu_3O_{7-\delta}$ but yet no reports have published regarding the ability of Al_2O_3 particles as pinning centre material. Until Mellekh et al. (2006) revealed that by using the Al_2O_3 nanoparticles with size about 50 nm, the J_C of $YBa_2Cu_3O_{7-\delta}$ was significantly improved. The improvement