UNIVERSITI PUTRA MALAYSIA

SYNTHESIS AND CHARACTERIZATION OF RARE EARTH BARIUM COPPER OXIDE [(RE1-xMx)Ba2Cu3O7-δ] VIA COPRECIPITATION AND ELECTROPHORETIC DEPOSITION METHODS

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SYNTHESIS AND CHARACTERIZATION OF RARE EARTH BARIUM COPPER OXIDE [$(RE_{1-x}M_x)Ba_2Cu_3O_{7-\delta}$] VIA COPRECIPITATION AND ELECTROPHORETIC DEPOSITION METHODS

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Chairman : Professor Zulkarnain Zainal, PhD

Faculty : Science

Preparation of superconducting material was usually carried out via the solid state reaction technique where oxides and carbonates metal precursors were mixed thoroughly followed by heat treatment. Alternatively, the materials can be prepared via the wet chemistry technique, in which higher homogeneity could be achieved due to initial mixture of cations at atomic scale. In this study, superconductors were prepared via co-precipitation technique from metal acetates which were reacted with oxalic acid as a precipitating agent to form metal oxalates.

Transmission Electron Microscopy (TEM) analysis shows that ultra fine powders of metal oxalates were obtained with the particle size in the range of 9 to 12 nanometers. X-ray diffraction (XRD) analyses confirmed the phases of the metal oxalates. Thermal decomposition and phase formation stages of samples were determined via Thermogravimetric (TG), Differential Thermal Analysis (DTA) and XRD analysis at different heat treatment temperatures.

Minute traces of $BaCuO_2$ phase was discernible in the X-ray diffraction patterns of sample obtained after sintering at 880 ^oC for 15 hours. By increasing the sintering temperature up to 970 ^oC, elimination of the $BaCuO_2$ phase in YNdBCO samples has succeeded. However, this phase remained in YGdBCO and NdGdBCO samples. Electrical resistivity measurement via 4-point probe technique shows zero resistance temperature, $T_{C(R=0)}$ and $T_{C\text{-onset}}$ are (86 K, 95 K), (88 K, 92 K) and (90 K, 98 K) for samples YNdBCO, YGdBCO and NdGdBCO, respectively.

In addition, preparation of superconductor coating were also carried out in this study. Numerous efforts have been reported in fabricating superconductors into certain shapes namely, wires, thin films and many more. The Electrophoretic Deposition (EPD) method is an attractive method where it is low cost, requires simple apparatus, large surface area with uniform thickness of deposits could be obtained and deposits could be designed according to the shape of substrate. In this study, the superconductor coatings were prepared via the EPD method. Several parameters such as colloidal suspension behaviour, voltage of deposition, grinding time, substrate and sintering conditions were investigated in order to obtain good quality coatings.

Colloidal suspension behaviour was investigated via Nanophox particle size analyzer. Smallest particle size distribution was ~ 15 nm by dispersing in acetone. Meanwhile, in order to enhance the deposition of the powders, particle size of the powders need to be reduced. After grinding the powders, deposition was improved and TEM analysis shows that deposited powders are in the range f 30 – 40 nm. To avoid the melting of silver substrates, sintering in argon atmosphere was carried out followed by slow cooling in

oxygen atmosphere. XRD analyses of deposited samples show that orthorhombic 123 phase was obtained YBCO, GdBCO and NdBCO samples however, for YBCO samples the orthorhombicity value was relatively low. Meanwhile, improvement in microstructure was achieved where more dense films were successfully obtained by performing the multi-deposition technique.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

PENYEDIAAN DAN PENGANALISISAN RARE EARTH BARIUM COPPER OXIDE [$(RE_{1-x}M_x)Ba_2Cu_3O_{7-\delta}$] MENGGUNAKAN KAEDAH PENGENAPAN CAMPURAN DAN PENGENAPAN ELEKTROFORETIK

Oleh

MOHD HANIFF BIN WAHID

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Penyediaan bahan superkonduktor biasanya dilaksanakan dengan menggunakan kaedah tindak balas struktur pepejal di mana prekursor logam dalam bentuk oksida dan karbonat dicampurkan bersama diikuti dengan pemanasan pada suhu tertentu. Sementara itu, kaedah alternatif untuk penyediaan bahan superkonduktor ini adalah melalui kaedah larutan kimia yang mana kaedah ini menawarkan produk yang lebih homogen disebabkan oleh campuran awal kation pada skala atom. Dalam kajian ini, bahan superkonduktor disediakan melalui kaedah pengenapan campuran di mana prekursor logam yang digunakan adalah logam asetat. Logam ini kemudiannya ditindakbalaskan dengan agen pengenapan iaitu asid oksalik untuk menghasilkan logam oksalat.

Hasil analisis mikroskop elektron transmisi TEM menunjukkan bahawa serbuk halus bersaiz 9 hingga 12 nanometer telah didapati. Manakala, pembelauan sinar X mengesahkan fasa sebatian yang dihasilkan. Penguraian termal dan peringkat

pembentukan fasa telah dikaji menggunakan kaedah termogravimetri TG, DTA dan pembelauan sinar X.

Setelah menjalani pensinteran pada suhu 880 ^{o}C selama 15 jam, didapati kewujudan fasa sekunder iaitu $BaCuO_2$ dalam jumlah yang sedikit terkandung di dalam bahan. Dengan meningkatkan suhu kepada 970 ^{o}C fasa sekunder tersebut berjaya dihilangkan dalam sebatian YNdBCO manakala fasa terbabit masih kekal dalam sebatian yang lain. Pengukuran rintangan elektrik melalui kaedah prob-4 menunjukkan nilai $T_{C(R=0)}$ and $T_{C\text{-}onset}$ adalah (86 K, 95 K), (88 K, 92 K) dan (90 K, 98 K) bagi sampel YNdBCO, YGdBCO dan NdGdBCO.

Selain daripada itu, penyediaan lapisan superkonduktor juga telah dilaksanakan. Pelbagai usaha telah dijalankan untuk memproses bahan superkonduktor ke dalam bentuk wayar, saput nipis, dan bermacam lagi. Kaedah pengenapan elektroforetik adalah menarik kerana kosnya yang rendah, memerlukan peralatan yang mudah, lapisan dapat memenuhi ruang yang besar serta memiliki ketebalan yang seragam boleh didapati, serta lapisan boleh dihasilkan dalam pelbagai bentuk mengikut bentuk substrat yang digunakan. Dalam kajian ini, lapisan superkonduktor disediakan melalui kaedah pengenapan elektroforetik. Pelbagai pemboleh ubah seperti sifat bahan terampai, voltan yang digunakan untuk pengenapan, masa yang digunakan untuk menghancurkan bahan, bahan substrat yang digunakan dan keadaan pensinteran telah dikaji untuk mendapatkan kualti lapisan yang terbaik.

Sifat larutan terampai telah dikaji menggunakan alat analisis partikel Nanophox. Serbuk telah didapati tersebar dengan baik dalam aseton berbanding dengan pelarut organik yang lain. Pensinteran bahan telah dijalankan dalam atmosfera argon bagi mengelakkan pencairan substrat. Hasil ujian pembelauan sinar-X ke atas sampel menunjukkan bahawa struktur Kristal ortorombik telah didapati bagi kesemua sampel. Namun, nilai ortorombisiti bagi sampel YBCO adalah agak rendah berbanding dengan nilai yang diperoleh dari JCPDS. Selain daripada itu, kualiti mikrostruktur lapisan telah dapat ditingkatkan di mana lapisan yang lebih padat telah berjaya diperoleh melalui kaedah pengenapan berulang.

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I certify that a Thesis Examination Committee has met on 22nd November 2010 to conduct the final examination of Mohd Haniff bin Wahid on his thesis entitled "Synthesis and Characterization of Rare Earth Barium Copper Oxide [$(RE_{1-x}M_x)Ba_2Cu_3O_{7-\delta}$] via Coprecipitation and Electrophoretic Deposition methods" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The committee recommends that the student be awarded the Master of Science.

Members of the Thesis Examination Committee were as follows:

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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.

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MOHD HANIFF BIN WAHID

Date:

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LIST OF ABBREVIATIONS

T_C	Critical temperature
YBCO	$YBa_2Cu_3O_{7-\delta}$
RE	Rare earth
J_C	Critical current density
RE-123	$REBa_2Cu_3O_{7-\delta}$
Y-123	$YBa_2Cu_3O_{7-\delta}$
REBCO	$REBa_2Cu_3O_{7-\delta}$
EPD	Electrophoretic deposition
COP	Co-precipitation
XRD	X-ray diffraction
TEM	Transmission Electron Microscope
BET	Brunauer-Emmet-Teller
TG	Thermogravimetric
DTA	Differential Thermal Analysis
XRF	X-ray Fluorescence
ICP	Inductive Coupled Plasma
SEM	Scanning Electron Microscope
JCPDS	Joint Committee on Powder Diffraction Standards
YGdBCO or YGd	$Y_{0.5}Gd_{0.5}Ba_2Cu_3O_{7-\delta}$
YNdBCO or YNd	$Y_{0.5}Nd_{0.5}Ba_2Cu_3O_{7-\delta}$
NdGdBCO or NdGd	$Nd_{0.5}Gd_{0.5}Ba_2Cu_3O_{7-\delta}$
$T_{C(R=0)}$	Critical temperature where resistance is totally zero

$T_{C(onset)}$	Critical temperature where superconducting transition starts

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