



UNIVERSITI PUTRA MALAYSIA

***IMPROVEMENT OF CRITICAL CURRENT DENSITY IN $YBa_2Cu_3O_{7-\delta}$
SUPERCONDUCTOR ADDED WITH Pr_2O_3 AND GRAPHENE OXIDE***

NUR ATIKAH BINTI BAHARUDDIN

FS 2018 112



**IMPROVEMENT OF CRITICAL CURRENT DENSITY IN $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$
SUPERCONDUCTOR ADDED WITH Pr_2O_3 AND GRAPHENE OXIDE**

By

NUR ATIKAH BINTI BAHARUDDIN

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,
in Fulfilment of the Requirements for the Degree of Master of Science**

January 2017

COPYRIGHT

All material contained within the thesis, including without limitation text, logos, icons, photographs, and all other artwork, is copyright material of Universiti Putra Malaysia unless otherwise stated. Use may be made of any material contained within the thesis for non-commercial purposes from the copyright holder. Commercial use of material may only be made with the express, prior, written permission of Universiti Putra Malaysia.

Copyright © Universiti Putra Malaysia



DEDICATION

To my father, mother,

Azizi, Izzati, Amin & my grandfather.



©

COPYRIGHT UPM

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

IMPROVEMENT OF CRITICAL CURRENT DENSITY IN $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ SUPERCONDUCTOR ADDED WITH Pr_2O_3 AND GRAPHENE OXIDE

By

NUR ATIKAH BINTI BAHARUDDIN

January 2017

Chairman : Associate Professor Mohd Mustafa Awang Kechik, PhD
Faculty : Science

The effect of nanoparticle praseodymium oxides (Pr_2O_3) and graphene oxides (GO) addition on the critical current density, J_c of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ was studied. Both nanoparticles added acted as pinning center to enhance the transport critical current density (J_c). $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superconductor was prepared by co-precipitation method with addition of Pr_2O_3 are 0.00, 0.02, 0.03, 0.04 and 0.05, while addition of GO are 0.0, 0.1, 0.2, 0.3, 0.4 and 0.5 weight percentage (wt.%). Co-precipitation process is based upon a simple control of the chemistry of the cation solution and precipitation with oxalic acid. The effect of nanoparticles addition on electrical superconducting properties (T_c and J_c), phase formation and structural properties, microstructure and surface morphology including the elemental distribution analysis were studied. $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ with Pr_2O_3 nanoparticle addition showed a maximal value of T_c at 82 K for 0.01 wt % addition and give smallest grain sizes 0.38 μm . However, J_c shows improvements in Pr_2O_3 addition of 0.03 wt.% with the highest J_c , 9678 A/cm^2 . $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ with GO nanoparticle showed an improvement value of T_c when GO added wt.% increase to 0.5 wt.%, 88 K. However, $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ added with GO give a vice versa results in J_c , which has a lower J_c than $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ with Pr_2O_3 nanoparticles addition. X-ray Diffraction (XRD), showed Y-123 phase exist in all samples which proves by the XRD and Energy Dispersive X-rays (EDX) profiling. Both nanoparticles added in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ show increased values of T_c and J_c towards the optimum wt. % of addition and decreased after further addition of nanoparticles.

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

PENAMBAHBAIKAN KETUMPATAN ARUS GENTING DALAM $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ SUPERKONDUKTOR DITAMBAH Pr_2O_3 DAN GRAPHENE OKSIDA

Oleh

NUR ATIKAH BINTI BAHARUDDIN

Januari 2017

Pengerusi : Profesor Madya Mohd Mustafa Awang Kechik, PhD
Fakulti : Sains

Kajian ke atas kesan nanopartikel praseodymium oksida (Pr_2O_3) dan graphene oksida (GO) dalam $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ superkonduktor ke atas arus ketumpatan genting J_c telah dijalankan. Kedua-dua nanopartikel bertindak sebagai pengepin fluks untuk meningkatkan pengangkutan ketumpatan arus genting (J_c). Serbuk $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ disediakan menggunakan kaedah se-pemendakan dengan menambah nanopartikel Pr_2O_3 ke dalam $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ dari peratusan berat (wt.%) 0.00, 0.01, 0.02, 0.03, 0.04, dan 0.05, manakala bagi penambahan nanopartikel GO dari 0.0, 0.1, 0.2, 0.3, 0.4 dan 0.5 peratusan berat. Proses se-pemendakan berlaku disebabkan oleh kawalan mudah kimia dalam larutan kation dan mendakan oksalid asid. Kesan nanopartikel dalam $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ terhadap sifat-sifat superkonduktor (T_c and J_c), fasa dan struktur, mikrostruktur dan morfologi termasuk analisis taburan unsur telah dikaji. $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ dengan nanopartikel Pr_2O_3 menunjukkan suhu genting, T_c pada 82 K bagi sampel 0.01%. Manakala, J_c menunjukkan perubahan yang baik dalam sampel 0.03% dengan ketumpatan arus genting yang tertinggi 9678 A/cm². Ini disebabkan aliran pengangkutan arus yang baik. $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ dengan nanopartikel graphene oksida (GO) memberi data J_c yang berlainan yang mana ketumpatan arus gentingnya lebih rendah berbanding sampel $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} + \text{Pr}_2\text{O}_3$. Analisis menunjukkan fasa Y-123 hadir dalam semua sampel yang mana dibuktikan dengan keputusan analisis pembelauan sinar-x, dan analisa profil tenaga penyerakan sinar-x (EDX). Kedua-dua nanopartikel yang ditambah ke dalam $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ menunjukkan nilai T_c dan J_c menurun apabila peratusan nanopartikel mencapai tahap optimum dalam $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$.

ACKNOWLEDGEMENTS

Praise to Allah. This dissertation would not have been possible without the guidance and the help of several individuals who in one way or another contributed and extended their valuable assistance in the preparation and completion of this study.

First and foremost, utmost gratitude to my supervisor Dr. Mohd Mustafa Awang Kechik for his unselfish and unfailing supports as my dissertation adviser, and always guides me in this research. A very special thanks to my Co-Supervisor, Assoc. Prof. Dr. Chen Soo Kien for always helping me with instrumentation and laboratory works. A big thanks to Prof. Dr. Abd Halim Shaari for his helps throughout this research. Also very big thanks to all Faculty of Science staff, En. Razak Harun, Pn. Norhaslinda Noorudin, Pn. Kamsiah Alias and many others for their great helps and contributions. Special thanks for Scheme Grant Research Assistant under Exploratory Grant Research Scheme (ERGS) with vot no 5527174, MyMaster under MyBrain from Ministry of Education, Malaysia and edu-loan from Majlis Amanah Rakyat (MARA) for financial support throughout my study.

A very special thanks to Assoc. Prof. Dr. Azhan Hashim (UiTM Pahang), for helping and guide me with J_c measurement using Four Point Probe while in UiTM Pahang, also special thanks to his post-graduate students Ms. Suazlina Mohd Ali for always helping me with all laboratory works while staying in UiTM. I would like to thanks my group members, Nur Nabilah Yusuf and Nur Hidayah Hapipi, for supporting me and I appreciate the discussions, ideas, memorable interactions and time spent throughout this research.

To my fellow friends, Nurul Auni Khalid, Wan Nur Wathiq Wan Jusoh, Noor Ariffuddin Jalani, Afiq Azri Zainuddin dan Yusnita Yusuf for always supports me through hell or high water.

Last but not least, I also would like to extend my thankfulness to the most precious persons in my life, my father and mother for all their moral support. I am deeply indebted to those responsible for making it possible for me to even consider a MSc and to those who helped me to achieve my goal.
Thank you a lots.

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee were as follows:

Mohd. Mustafa Awang Kechik, PhD

Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Chen Soo Kien, PhD

Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

Azhan bin Hashim, PhD

Professor, Ts.
Faculty of Applied Sciences
Universiti Teknologi MARA Pahang
Pahang, Malaysia
(Member)

ROBIAH BINTI YUNUS, PhD

Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 08 March 2018

Declaration by graduate student

I hereby confirm that:

- this thesis is my original work;
- quotations, illustrations and citations have been duly referenced;
- this thesis has not been submitted previously or concurrently for any other degree at any institutions;
- intellectual property from the thesis and copyright of thesis are fully-owned by Universiti Putra Malaysia, as according to the Universiti Putra Malaysia (Research) Rules 2012;
- written permission must be obtained from supervisor and the office of Deputy Vice-Chancellor (Research and innovation) before thesis is published (in the form of written, printed or in electronic form) including books, journals, modules, proceedings, popular writings, seminar papers, manuscripts, posters, reports, lecture notes, learning modules or any other materials as stated in the Universiti Putra Malaysia (Research) Rules 2012;
- there is no plagiarism or data falsification/fabrication in the thesis, and scholarly integrity is upheld as according to the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) and the Universiti Putra Malaysia (Research) Rules 2012. The thesis has undergone plagiarism detection software

Signature: _____ Date: _____

Name and Matric No: Nur Atikah binti Baharuddin, GS38076

Declaration by Members of Supervisory Committee

This is to confirm that:

- the research conducted and the writing of this thesis was under our supervision;
- Supervision responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) were adhered to.

Signature: _____

Name of Chairman
of Supervisory
Committee:

Associate Professor
Dr. Mohd. Mustafa Awang Kechik

Signature: _____

Name of Member
of Supervisory
Committee:

Associate Professor
Dr. Chen Soo Kien

Signature: _____

Name of Member
of Supervisory
Committee:

Professor Ts.
Dr. Azhan bin Hashim

TABLE OF CONTENTS

	Page
ABSTRACT	i
ABSTRAK	ii
ACKNOWLEDGEMENTS	iii
APPROVAL	iv
DECLARATION	v
LIST OF TABLES	x
LIST OF FIGURES	xi
LIST OF ABBREVIATIONS	xiv
CHAPTER	
1 INTRODUCTION	1
1.1 Introduction of Superconductor	1
1.2 History of Superconductor	2
1.3 Problem statement	3
1.4 Objectives	4
1.5 Outline of Thesis	4
2 THEORY AND LITERATURE REVIEW	5
2.1 Introduction	5
2.2 Theory of Superconductor	5
2.2.1 Fundamental Properties of Superconductor	5
2.2.2 The Meissner Effect	6
2.2.3 Type I Superconductors	9
2.2.4 Type II Superconductors	9
2.2.5 BCS (Bardeen Cooper-Schrieffer) Theory	10
2.2.6 Pure YBa ₂ Cu ₃ O _{7-δ} perovskite	10
2.3 Literature Review	11
2.3.1 Nanoparticle addition on YBCO	11
2.3.2 Co-precipitation method	16
3 METHODOLOGY	18
3.1 Introduction of Experimental Details	18
3.2 Synthesis of Yttrium Barium Copper Oxide (YBCO)	18
3.2.1 Mixture Ratio	18
3.2.2 Co-Precipitation (CO-P) Process	18
3.2.3 Calcination of YBCO precursor	19
3.2.4 Praseodymium oxide (Pr ₂ O ₃) and Graphene oxide (GO) nanoparticles and palletisation	20
3.2.5 Final sintering	20
3.3 Characterization of sample	21
3.3.1 Thermogravimetry Analysis (TGA)	21
3.3.2 Electrical Resistance at various temperature	21
3.3.3 X-Ray diffraction (XRD)	22

3.3.4	Scanning Electron Microscope (SEM)	23
3.3.5	Energy-dispersive X-ray spectroscopy (EDX)	23
4	NON-ADDED YBCO AND YBCO WITH ADDITION OF Pr₂O₃	25
4.1	Introduction	25
4.1.1	Thermogravimetric analysis (TGA) of YBCO	25
4.2	Characterization of YBa ₂ Cu ₃ O _{7-δ}	26
4.3	Praseodymium oxides (Pr ₂ O ₃) as nanoparticle in YBCO system	30
4.3.1	X-Rays Diffraction Analysis (XRD)	31
4.3.2	Micrograph analysis using (SEM)	33
4.3.3	Energy Dispersive X-ray (EDX) Analysis	36
4.3.4	Critical temperature analysis T_c	39
4.3.5	Critical current density J_c analysis for YBCO + wt. % Pr ₂ O ₃	41
5	YBCO WITH ADDITION OF GRAPHENE OXIDES (GO)	44
5.1	Characterization of YBa ₂ Cu ₃ O _{7-δ} + wt% of GO	44
5.2	Graphene Oxides as Nanoparticle in YBCO	44
5.2.1	X-ray Diffraction Analysis (XRD)	45
5.2.2	Micrograph analysis using (SEM)	48
5.2.3	Energy Dispersive X-ray Analysis	51
5.2.4	Critical temperature, T_c analysis YBCO +wt.% of GO	54
5.2.5	Critical current density (J_c) analysis for YBCO +wt. % GO	55
6	CONCLUSIONS AND SUGGESTIONS	57
6.1	Conclusions	57
6.2	Suggestions	58
	REFERENCES	59
	APPENDICES	65
	BIODATA OF STUDENT	70
	LIST OF PUBLICATIONS	71

LIST OF TABLES

Table		Page
2.1	The alloys of Type II Superconductor	9
3.1	Metal acetates used for CO-P	18
4.1	Abbreviation and description of bulk samples of YBCO + Pr ₂ O ₃ wt.% (0.00 – 0.05)	30
4.2	Unit cell lattice parameters of <i>a</i> , <i>b</i> and <i>c</i> axis and unit cell volume for YBa ₂ Cu ₃ O _{7-δ} + different wt% of Pr ₂ O ₃ (0.00, 0.01, 0.02, 0.03, 0.04 and 0.05)	32
4.3	Average particle size for YBCO + wt. % of Pr ₂ O ₃ samples from SEM micrograph	35
4.4	The ratio of Y-123 phase from different nanoparticles of Pr ₂ O ₃	39
4.5	Critical temperature <i>T_c</i> for all samples	41
5.1	Abbreviations for bulk samples	45
5.2	Lattice parameter and unit cell volume calculated by XRD spectrum for all YBCO + different wt.% of GO	48
5.3	Average particle size for YBCO + wt.% of GO samples from SEM micrograph	50
5.4	The ratio of Y-123 phase from different nanoparticle of GO	51

LIST OF FIGURES

Figure		Page
1.1	The chronology of discoveries of superconductors	3
2.1	Graph of resistance against temperature. The graph shows the difference between a non-superconductor metal and a superconductor metal. For superconducting material, the resistivity suddenly dropped to zero as the temperature approaching the critical temperature, T_c	7
2.2	Diagram illustrate the phenomenon of Meissner effect as in Figure 2.3. The magnetic field lines being expelled by the superconducting material and hence the magnet to float above it	7
2.3	The levitating of YBCO superconductor Meissner effect taken in Laboratory of Superconductor and Thin Film, Faculty of Science, UPM	8
2.4	The Meissner Effect. When temperature is greater than the critical temperature, T_c , the magnetic field will penetrate inside the material (left) whereas as the material cooled below the critical temperature, the material will expel the magnetic field and become a superconducting material (right)	8
2.5	Y-123 has the orthorhombic crystal structure. It has one yttrium atom at the body centred in between the Cu-O planes, two barium atoms at the body centred, copper atoms located at the edge of every lattice with oxygen atoms in between them	11
2.6	Resistivity graph versus temperature for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. As the concentration of Pr increase, the T_c monotonically decrease	13
2.7	Shows the self-field J_c with different wt.% of nanoparticle addition to get a better comparison of J_c . The J_c values indicates that the nanoparticle added samples has better performance than pure sample, with J_c of the nano powder added sample improved up to a factor of ~ 3	14
2.8	TEM image for Co_3O_4 with average grain size of 30 nm	15
2.9	X-ray diffraction patterns for pure MgB_2 , GO-added, rare-earth added MgB_2 , denoted as MBR (R = La, Sm, Eu, Gd, Tb and Ho) series and GO and rare-earth added MgB_2 , denoted as MBRG (R = La, Sm, Eu, Gd, Tb and Ho) series of samples. XRD results show the presence of rare earth impurity phase in superconductor, which may act as strong pinning centers in the samples	15

2.10	This is the general idea on how to prepare the Y-123 precursor by using co-precipitation method. Basically, ingredients in beaker A will mixed up with ingredients in beaker B before we put the mixture in ice bath. After some time of mixing in ice bath, the mixture will go through filtration process for 5 hours before we get a blue precursor. Then, the blue precursor will be ready for drying process before going through calcination and sintering process	17
3.1	Heating profile for 1st and 2nd calcinations process. The powder was calcined from room temperature to 900°C with heating rate of 2°C/min and hold for 12 hours before cooled back to room temperature with cooling rate of 1°C/min	20
3.2	Heating profile for sintering process. The sample undergo heating process on 920°C for 15 hours then been cooling down to 680°C for another 8 hours before been cooling to room temperature	21
3.3	Shows a typical X-ray Diffraction diagram for analyzing the phase and structure of the samples	24
3.4	Shows a schematic diagram of Scanning Electron Microscopy (SEM)	24
4.1	Graph of weight loss vs. mass (TGA)	26
4.2	The resistance vs. temperature for YBCO sample	28
4.3	X-ray diffraction patterns of pure YBCO	28
4.4	a) Micrograph structure for pure YBCO. b) EDX spectrum from spectrum 2 shows Y-123 as the major phase with 1.00: 1.86: 2.62	29
4.5	SEM image of pure YBCO with an average grains size 1.02 μm	29
4.6	The micrograph of Pr ₂ O ₃ nanoparticle taken using FESEM at 2500x magnification. The particle of Pr ₂ O ₃ agglomerates because the texture of the powder very fine. The average grains size of Praseodymium which is ~ 0.733 μm	31
4.7	X-ray diffraction patterns of YBa ₂ Cu ₃ O _{7-δ} + different wt.% of Pr ₂ O ₃ (0.00, 0.01, 0.02, 0.03, 0.04 and 0.05)	32
4.8	SEM images showing YBCO samples with different weight percentage of Pr ₂ O ₃ . a) 0.00 wt. % Pr ₂ O ₃ , b)0.01 wt. % of Pr ₂ O ₃ , c) 0.02 wt. % of Pr ₂ O ₃	34
4.9	SEM images showing YBCO samples with different weight percentage of Pr ₂ O ₃ . d) 0.03 wt. % of Pr ₂ O ₃ , e) 0.04 wt. % of Pr and f) 0.05 wt. % of Pr ₂ O ₃ . All samples were taken at 5000x magnification except P0 samples which is 2000x	35

4.10	Surface morphology of a) P0, b) P1 and c) P2 with EDX spectrum. EDX spectrum on the right side shows the element detected from micrograph	37
4.11	Surface morphology of d) P3, e) P4 and f) P5 with EDX spectrum. EDX spectrum on the right side shows the element detected from micrograph	38
4.12	Critical temperature, T_c , for all samples from 0.01 wt. % till 0.05. wt % of Pr_2O_3	40
4.13	J_c value for all addition of Pr_2O_3 at 30 K. At 30 K, the J_c value for P0, P1, P2, P3, P4 and P5 are 11.03 A/cm ² , 710.00 A/cm ² , 684.07 A/cm ² , 9678 A/cm ² , 141.61 A/cm ² and 43.87 A/cm ² respectively. The graph shows an improvement of J_c in addition of P3 (0.03) wt.% of Pr_2O_3 in YBCO which 9678 A/cm ²	42
4.14	Graph shows the combination of J_c for P0, P1, P2, P3, P4 and P5 temperature 30 K, 50 K, 60 K, 70 K, 77 K and 80 K. Clearly can be seen, P3 has the highest J_c among all, follow with P1 and P2 compare to P4 and P5	43
5.1	The micrograph of Graphene oxides sheet taken using FESEM at 220x magnification	45
5.2	XRD diffraction patterns of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ + different wt.% of GO at	47
5.3	SEM images of YBCO + wt. % of Graphene oxides (GO) at 2000x magnification. a) 0.0 wt. % GO, b) 0.1 wt. % of GO, c) 0.2 wt. % of GO	49
5.4	SEM images of YBCO samples with different wt. % of Graphene oxides (GO) at 2000x magnification. d) 0.3 wt. % of GO, e) 0.4 wt. % of GO, and f) 0.5 wt. % of GO	50
5.5	a) Surface morphology for P0 with EDX spectrum. b) Surface morphology for G1 with EDX spectrum. c) Surface morphology for G2 with EDX spectrum	52
5.6	d) Surface morphology for G3 with EDX spectrum. e) Surface morphology for G4 with EDX spectrum. f) Surface morphology for G5 with EDX spectrum	53
5.7	Critical temperature T_c for all samples from 0.1 wt % of GO till 0.5 wt % of GO	55
5.8	The critical current density, J_c , for all YBCO + wt%. of Graphene oxides from (0.1, 0.2, 0.3, 0.4 and 0.5)	56

LIST OF ABBREVIATIONS

a.u	Arbitrary Unit
AMU	Atomic mass unit
APC	Artificial Pinning Centre
BaCuO	Barium Copper Oxide
BCS	Bardeen-Cooper-Schrieffer
CO-P	Co-precipitation
ξ	Coherence length
T_c	Critical temperature
J_c	Critical current density
EDX	Energy Dispersive X-ray
FESEM	Field Emission Scanning Electron Microscopic
GO	Graphene Oxide
HTSC	High Temperature Superconductor
K	Kelvin
x	Magnification
MRI	Magnetic Resonance Imaging
Pr_2O_3	Praseodymium Oxide
λ	Penetration depth
RE	Rare Earth
SEM	Scanning electron microscope
SQUID	Superconducting Quantum Interference Devices
TGA	Thermogravimetric analysis
wt.%	Weight percentage
XRD	X-ray diffractometer
YBCO	Yttrium Barium Copper Oxide,
Y-123	$\text{YBa}_2\text{Cu}_3\text{O}_{7.5}$
Y-124	$\text{YBa}_2\text{Cu}_4\text{O}_8$

CHAPTER 1

INTRODUCTION

1.1 Introduction of Superconductor

The discovery of High Temperature Superconductor (HTSC) ceramic materials by Bednorz and Muller in 1986 has brought the possibility of superconductor applications at liquid nitrogen temperature. A great attention has been devoted to search for ways to produce affordable flexible conducting HTSC wires with high current density, which can be used for many applications (Zhou *et al.*, 2008). Applications of superconductor can be divided into two categories; large scale and small scale. Large scale application includes high speed train, magnetic energy storage, magnetic resonance imaging (MRI) for medical applications and high energy physics instruments. Small scale application includes Josephson devices, Superconducting Quantum Interference Devices (SQUIDS), microwave devices and resonators (Abd-Shukor, 2004).

Superconductor is a material that can conduct electricity without any resistance. There are no losses in form of energy that can be released from the material until it reached certain temperature, or critical temperature T_c which the material becomes superconductive (Frank, 2002). This phenomenon occurred where certain materials exhibit when it is cooled down below the T_c , this is known as the Meissner effect. There are two important properties of superconductors, which are zero dc resistivity and perfect diamagnetism. Both can be used to enhance the performance of many devices such as large scale application, MRI.

To promote industrial application, the relationship between the density of the critical current density J_c and the composition, processing and microstructural features of polycrystalline $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) ceramic should be understood. Many researches have been conducted to improve the YBCO system in order to achieve higher T_c , and in higher J_c . In the preparation of YBCO superconductor, nanoparticle is added in the YBCO powder as a pinning mechanism (Khalil, 2001; Matsumoto *et al.*, 2004; Mizan & Kong, 2012).

Pinning mechanism will act as flux pinning in YBCO system, to control the flux creeps inside the grains. We believe, as we control the flux flow, the properties of superconductor will be enhanced (Orbital, 2004; Haugan, Barnes, Wheeler, 2004). There are many examples of pinning mechanism such as silicon, germanium, cadmium, samarium, selenium and neodymium. However, in the midst of searching the best nanoparticles to improve T_c , we found that the addition of rare earth materials could improve the properties of superconductor. The control of both the composition and preparation routes is known to be the key factor for the regulation of the flux pinning in the superconducting ceramics (Raouf, 2005).

1.2 History of Superconductor

Superconductivity was discovered on 1911 by Heike Kamerlingh Onnes, who was studying the resistance of solid mercury at cryogenic temperatures. He discovered that the electrical resistivity of the solid mercury suddenly dropped to zero when it was cooled down to temperature below 4.2 K, which is the boiling point of liquid helium (Kisa, 1997). Onnes noticed that if he turned off the voltage below 4.2 K, the current continued to flow without loss. The liquid helium was first produced by Onnes in 3 years before liquefying the helium. This event marked the beginning of discovery of superconductor (Ireson, 2012).

Later, many metallic elements were found to exhibit superconductivity at very low temperature. In 1930, the highest critical temperature of all pure metal is Nb, with the T_c of 9.2 K. One of the distinct properties of the superconducting state in superconductors is perfect diamagnetism or Meissner effect, which was discovered by Meissner and Ochsenfeld in 1933. One year later, Fritz and Heinz London proposed the London model which explained the Meissner effect and predicted the penetration depth λ . Ginzburg-Landau theory was proposed and published seventeen years later (Ireson, 2012).

Alexei Abrikosov (1957) showed that superconductor can be classified into Type-I and Type-II superconductor. Then, John Bardeen, Leon Cooper and Robert Schrieffer proposed the BCS theory of superconductor. BCS theory explained about the Cooper pairs that usually occurred in conventional and Type-I superconductor. It was then followed by Brian Josephson, where he postulated a quantum tunneling effect which is known as Josephson effects (Das, 2009; Paul Leonard Bach, 2011).

The development of superconductivity came to a breakthrough in the year 1987, when the research groups in Alabama and Houston coordinated by K. Wu and Paul Chu discovered $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ceramics with $T_c = 92$ K. Later, more high- T_c cuprates oxides like $\text{Bi}_2\text{Ca}_2\text{Sr}_2\text{Cu}_3\text{O}_{10}$ with $T_c = 110$ K and $\text{Tl}_2\text{Ca}_2\text{Ba}_2\text{Cu}_3\text{O}_{10}$ with $T_c = 125$ K were discovered (Segrè, 2001).

$\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ is one of the most famous copper based superconductors because it is the first material that achieved the critical temperature higher than previously found superconductor as shown in Error! Reference source not found.. It also has the critical magnetic field of 300 T. It was an important discovery because the critical temperature of $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ is above the boiling point of liquid nitrogen, 77 K, which is a cheaper and easier to handle coolant as compared to liquid helium.

The superconducting material will be able to levitate in the air without any contact to neither solid nor liquid medium. The most important factors concerning the high-temperature superconductors for technical applications are their capacity in carrying

vast current densities (Grant, 2001). The value of critical current density, J_c which is higher than 10^6 A/cm at liquid nitrogen temperature (77 K) has been achieved. Moreover, there is no strong magnetic field that can destroy superconductivity (Tavana & Akhavan, 2010).

Many research groups around the globe are doing continuous experiment in seeking for new superconducting materials with higher transition temperatures. Scientists and experts hope that they will discover another family of superconducting material which will simplify the task of explaining how these materials display the properties of superconductivity.

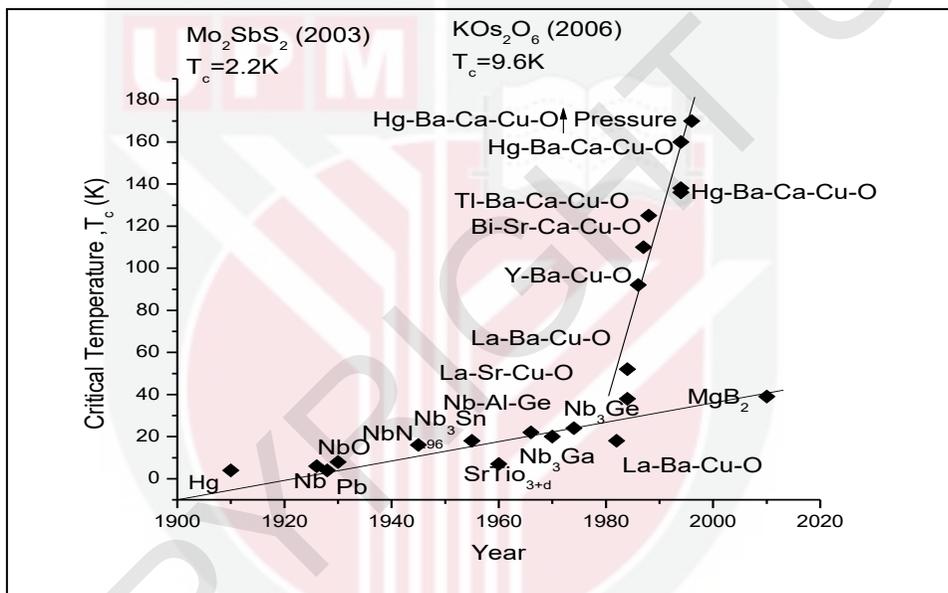


Figure 1.1 : The chronology of discoveries of superconductors (Cheong, 1996)

1.3 Problem statement

Previous study proved that flux pinning is very important in HTSC where supercurrent can flow without any energy dissipative. However, since the discovery of cuprates (copper-oxide-based-materials) superconductor in early 1980 (Kim, 2007), many researches have been focusing on enhancing the superconductor properties by adding nanoparticles into system. In order to produce ultrafine powder of precursor of superconductor, co-precipitation method was chosen due to the low reaction temperature. Other than that, nanoparticles of praseodymium oxide (Pr_2O_3) and graphene oxide (GO) are added to $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. Nanoparticles were selected due to their own properties which may increase critical current density, J_c .

1.4 Objectives

Hence, this study focuses on the following objectives:

- 1) To synthesize a good quality of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ with ultrafine grain size by using the co-precipitation method.
- 2) To study the effect of nanoparticle addition of praseodymium oxide (Pr_2O_3) and graphene oxide (GO) by adding different weight percentage on critical temperature, T_c and critical current density, J_c , of superconducting $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$.
- 3) To investigate the effect of nanoparticle addition on physical properties (phase, structure and morphology).

1.5 Outline of Thesis

The thesis is outlined as follows; Chapter 1 consists of general introduction on the research, history of superconductivity, problem statement and the objectives of research. Chapter 2 highlights the basic theory and literature review of basic properties of superconductor, crystal structure and the effect of the nanoparticles addition for various types of elements on the superconducting properties of YBCO. Details of the sample preparation and experimental methods to perform the measurements are being discussed in Chapter 3. While in Chapter 4 and Chapter 5, the experimental data and results obtained for each nanoparticles added as well as discussions are presented. Finally, the relation between phase, structure and superconducting properties is concluded and suggestions for future works are given in Chapter 6.

REFERENCES

- A.Mellekh, M.Zouaoui, F.Ben Azzouz, M.Annabi, & Salem, M. B. (2006). Nano- Al_2O_3 particle addition effects on $\text{YBa}_2\text{Cu}_3\text{O}_7$ superconducting properties. *Solid State Communications*, 140(6).
- Abu Ziad, S. (2010). New Investigation for T_c depression by Ca in $\text{Y}_{1-x}\text{Ca}_x\text{O}_{123}$ superconducting systems. *Physica C; Superconductivity*, 470(17-18), 659–668.
- Alechu, G. (2001). The Morphology of Some YBCO Superconductor Materials. *Metal*
- Aleksandr Dubov. (2015). Finite size effects in superconducting nanocomposites. *Lappeenranta University Of Technology*
- Annunziata, G. (2010). Transport And Proximity Effect In Unconventional Ferromagnet / Superconductor Heterostructures. *Università degli Studi di Salerno*
- Azman, N. Jannah, Abdullah, H., & Abd-shukor, R. (2014). Transport Critical Current Density of $(\text{Bi}_{1-x}\text{Pb}_x)\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ Ceramic Superconductor with Different Nanosized Co_3O_4 Addition, *Advance in Condensed Matter Physics*
- Bhargava, Mackinnon, Yamashita (1995), Bulks manufacture of YBCO powders by coprecipitation, *Physica C Superconductivity* 241(1): 53-62.
- Babu, N. H., Lo, W., Cardwell, D. A., & Shi, Y. H. (2000). Fabrication and microstructure of large grain Nd–Ba–Cu–O, *Superconductor Science Technology* 13, 468–472.
- Babu, N. H., Shi, Y., Pathak, S. K., Dennis, A. R., & Cardwell, D. A. (2011). Developments in the processing of bulk (RE) BCO superconductors. *Physica C: Superconductivity and Its Applications*, 471(5-6), 169–178.
- Baharlooie, E., & Nazifi, R. (2014). Critical Current Density Enhancement in MTG YBCO Doped with Y. *Iranian Conference on Electrical Engineering*, 488–490.
- Benzi, P., Bottizzo, E., & Rizzi, N. (2004). Oxygen determination from cell dimensions in YBCO superconductors. *Journal of Crystal Growth*, 269, 625–629.
- Burns, G. (1992) High Temperature Superconductivity.
- Bhargava, A., Mackinnon, I. D. R., Yamashita, T., & Page, D. (1995). Bulk manufacture of YBCO powders by coprecipitation, *Physica C*, 241, 53–62.
- Cao, H., Hu, Rui., Li, Jinshan., Kou, H., & Zhou, L. (2008). Analysis on the interface stability and morphology evolution rules of the YBCO crystal growth during the unidirectional solidification, *Journal of Alloy and Compounds*, 462, 428–431.

- Cheong, P. T. (1996). Characterisation and Application of Praseodymium Doped YBCO Compounds. *University of Hong Kong*
- Cheung, Y. L., Maher, E. F., Abell, J. S., Jones, I. P., & Tse, Y. Y. (2007). Microstructural study of a YBCO multilayer coated conductor cylinder. *Superconducting Science and Technology*, 20, 511–515.
- Das, S., Bernhard, C., & Varma, G. D. (2015). Effect of combined addition of graphene oxide and citric acid on superconducting properties of MgB₂. *Physica C Super Conductivity*, 509, 49–55.
- Das, T. (2009). A model of coexistence of antiferromagnetism and superconductivity in electron- and hole-doped cuprates. *Northeastern University*
- Dangerfield, M. (2011). "Defrosting the Secrets of Superconductivity." Retrieved from <http://www.cankler.com.au/category/protoscience-2/>.
- Dekkers, J. M., Rijnders, G., Harkema, S., Smilde, H. J. H., Hilgenkamp, H., Rogalla, H., & Blank, D. H. A. (2003). Monocrystalline YBa₂Cu₃O_{7- δ} thin films on vicinal SrTiO₃ 001 substrates, *Applied Physics Letters* 83(25), 5199–5201.
- Drandova, G. I. (2001). Nmr Investigations In Copper-Oxide Chain Compounds And High-Tc Superconductors. *The University of Texas*
- Eun, J. (2012). Theory of Quantum Oscillations in Cuprate Superconductors. *University of California*
- Frank J. (2002). The New Superconductors
- Fuente, J. de La. (cited 2015) Graphene Oxides-What is it? *Graphenea*. Retrieved from <http://www.graphenea.com>
- Gill, R., & Singh, P. (2005). Study of depression of critical temperature of praseodymium doped high T_c cuprates, 43, 977–979.
- Giri, R., Awaria, V.P.S.Singh, H, K, Tiwari, R.S, Srivasdava, O.N.Gupta, A.Kumaraswamy, B.V and Kishan, H. (2005). Effect of Ca doping for Y on structural/microstructural and superconducting properties of YBa₂Cu₃O_{7- δ} *Physica C:Superconductivity*, 419 (3-4), 101–108.
- Grant, P. M. (2001). The critical current of superconductors: an historical review. *Low Temperature Physics*, 27(9).
- Grinenko, V. A. (2007). Upper critical field of cellular magnesium diboride. *JETP Letters*, 85(12), 622–625.
- Hafiz, M. (2015). Effect of Nanosized NiF₂ Addition on the Transport Critical Current Density of Ag-Sheathed (Bi_{1.6}Pb_{0.4})Sr₂Ca₂Cu₃O₁₀ Superconductor Tapes. *Advanced in Materials Science and Engineering*

- Harrison, B. C., Kell, J. W., Barnes, P. N., Haugan, T. J., Rane, M. V., & Ramos, F. (2012). Pr Doped Ybco Films Produced By Pulsed Laser Deposition (Postprint) Propulsion Directorate. *Material Research Society*
- Hopkins, S. C. (2007). Optimisation, Characterisation and Synthesis of Low Temperature Superconductors by Current-Voltage Techniques. *Sidney Sussex College*
- Ireson, G. (2012). Discovering Superconductivity. *WILEY*
- K. S. B. De Silva, X. Xu, W. X. Li, Y. Zhang, M. Rindfleisch, and M. T. (2011). Improving Superconducting Properties of MgB₂ by Graphene Doping. *IEEE Transactions On Applied Superconductivity*, 21(3), 2686–2689.
- Kang, J., Haffner, S., Olson, C. G., Kim, J. H., Maple, M. B., Kwon, S. K., & Min, B. I. (2003). Angle-resolved valence-band photoemission study of the quenched superconductors : Y_{1-x}Pr_xBa₂Cu₃O_{7-d}, *Physica B*, 328, 15–19.
- Kechik, A. (2010). Improvement of Critical Current Density in YBa₂Cu₃O_{7-δ} Films with Nano- Inclusions, *University of Birmingham*
- Khaled M. Elsabawy, M.M.A.-S. (2012). Structure Stability Of Optimally-Praseodymium, *Structure B-Research Paper*
- Khalil, A. E. (2001). Morphology of defects and the transport properties of high-T_c superconductors. *Physica C*, 353, 133–140.
- Kim, S. II. (2007). The Critical Current Density of YBa₂Cu₃O_{7-x} Coated Conductors. *University of Wisconsin – Madison*
- Kisa, P. (1997). Influence Of Defects On Critical Parameters In High Temperature Superconductors. *University of Belgrade*
- Krey. (2006). The Superconductor YBCO. Retrieved from <http://www.kreynet.de/asc/ybco.html>
- L.S Uspenskaya, I. N. (2002). Effect of Mesoscopic Inhomogeneities on the Critical Current of Bulk Melt-textured YBCO. *Solid State Physics*, 5(211), 1–9.
- M.M.Kamarulzaman, S.A.Halim, M.I. Adam, M.A.Faisal. (2010). Structure and superconductivity in YBCO with additives of Nano Sm₂O₃., 1250 (1), 492–495.
- M.Mujaini, S.Y.Y, I. Hamadne, and R.A.S. (2009). Synthesis of YBCO High-Temperature Superconductor. *AIP*
- Matsumoto, K., Horide, T., Osamura, K., Mukaida, M., Yoshida, Y., Ichinose, A., & Horii, S. (2004). Enhancement of critical current density of YBCO films by introduction of artificial pinning centers due to the distributed nano-scaled

Y2O3 islands on substrates. *Physica C: Superconductivity*, 412-414, 1267–1271.

Mennema, S. (2006). Normal state properties of high-angle grain boundaries in $Y_{1-x}Ca_xBa_2Cu_3O_{7-d}$. *University of Cambridge*

Nikolo, M. (1994). Superconductivity: A guide to alternating current susceptibility measurements and alternating current susceptometer design. *American Journal Physics*, 63(1).

R.Abdul-Shukor, I.Kong, E.L.Lim, N.A.Mizan, H.A.Alwi, M.H.Jumali, W.Kong (2012). Enhanced Critical Current Density of FeF₂ Added YBa₂Cu₃O_{7-δ}, *Journal Superconductor* 957–960.

Mouganie, T. (2005). The ink-jet printing of an all-chemical solution deposition process for YBa₂Cu₃O_{7-δ} coated conductors. *University of Cambridge*

Orbital, H. F. (2004). Addition of nanoparticle dispersions to enhance flux pinning of the, *Nature*, 430(August), 867–870.

P.F Henning, G.CaO, J.E.Crow, W.O.Puttika, P. J. H. (1995). Thermal Conductivity of Zn,Pr and Tb doped YBCO Single Crystal: Theory and Experiment. *Journal of Superconductivity*, Vol 8(No 4).

Parinov. (2007). Microstructure and Properties of High-Temperature Superconductors. *Springer*

Pathak, L. C., Mishra, S. K., Das, S. K., Bhattacharya, D., & Chopra, K. L. (2001). Effect of sintering atmosphere on the weak-link behaviour of YBCO superconductors, 351, 295–300.

Paul Leonard Bach. (2011). Electronic Transport Measurements In The Electron-Doped High-Temperature Superconductor Pr_{2-x}Ce_xCuO, *University of Maryland*

Pong, I. (2008). Phase and Microstructure Development during Multi-stage Heat Treatment of “ Internal Tin ” Ti-doped Nb₃Sn Superconducting Multifilamentary Wire Ian Pong St John ’ s College Applied Superconductivity and Cryoscience Group Department of Materials Science.

Poonam Rani, A. P. and V.P.A. (2012). High field magneto-transport study of YBCO:Ag_x (0.00-0.20). *National Physical Laboratory*

R.Abd-Shukor. (2004). Introduction to Superconductivity. *Universiti Pendidikan Sultan Idris*

Raouf, D. N. (2005). Bulk Properties of YBa₂Cu₃O₇ Superconducting Materials, *Journal of Applied Physics*, 1(2), 19–22.

- Rejith, P. P., Vidya, S., & Thomas, J. K. (2015). Influence of $\text{YBa}_2\text{HfO}_{5.5}$ - “ derived secondary phase ” on the critical current density and flux-Pinning force of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thick films. *Cryogenics*, 72, 1–8.
- Sheahen, T. P. (1994). Introduction to High-Temperature Superconductivity. New York, London, *Plenum Press*
- S. Stankovich, D. A. Dikin, R. D. Piner, K. A. Kohlhaas, A. Kleinhammes, Y. Jia, Y. Wu, S. B. T. N. and R. S. R. (2007). Synthesis of Graphene-Based Nanosheets via Chemical Reduction of Exfoliated Graphite Oxide. *Carbon*, Vol. 45, 1558–1565.
- Sahoo, M., & Behera, D. (2012). Electrical transport Property, 2(9), 1–6.
- Salleh, K. (2011). Influence Of Heat Treatments On Electrical Properties And Microstructure Of Porous Ybco Superconductor, *Universiti Teknologi MARA*
- Sato Kazuyoshi, O. S. (2009). Synthesis of NiO / YSZ Co-precipitation Method Nanocomposite Particles using Co-Precipitation Method. *Transactions of JWRI*, 38.
- Segrè, G. P. (2001). Pump Probe Spectroscopy of Quasiparticle Dynamics in Cuprate Superconductors. *University Of California*
- Sinclair, J. W. (2011). An Investigation of Pinning Landscapes with Engineered Defects : Contact-free Critical Current Density Measurements. *University of Tennessee*
- Sun, Q. J., Wang, H. S., Wang, H. M., Deng, L. W., Hu, Z. W., Gao, B., Li, Q. (2014). Electronic transport transition at graphene / $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ junction, 102602, 3–8.
- T.Haugan, P.N.Barnes, R.Wheeler, F. M. & M. S. (2004). Addition of nanoparticles dispersions to enhance flux pinning of the YBCO superconductor. *Nature*, 430, 867–870.
- Tassin, P., Koschny, T., Kafesaki, M., & Soukoulis, C. M. (2012). A comparison of graphene , superconductors and metals as conductors for metamaterials and plasmonics, *Iowa State University*, 1–19.
- Tavana, A., & Akhavan, M. (2010). How T_c can go above 100 K in the YBCO family. *European Physical Journal B*, 73(1), 79–83.
- Thomas, A. E. (1994). Synthesis and characterization of high temperature cuprate superconductors. *Temple University*
- Venkataraman Thangadurai, Robert A. Huggins, W. W. (2001). Mixed ionic-electronic conductivity in phases in the praseodymium oxide system. *Journal of Solid State Electrochemistry*, 5(7), 531–537.

Wang, X. (2011). Superconducting Properties of Graphene Doped Magnesium Diboride, *Faculty of Engineerings-Papers*, 201–218.

Xu, X., Dou, S. X., Wang, X. L., & Kim, J. H. (2010). Graphene doping to enhance flux pinning and supercurrent carrying ability in magnesium diboride superconductor. *Superconducting & Electronic Materials*, 1–12.

Yin, P. T., Shah, S., Chhowalla, M., & Lee, K. (2014). Design , Synthesis , and Characterization of Graphene–Nanoparticle Hybrid Materials for Bioapplications. *Chemical Reviews*

Zhou, H., Maiorov, B., Wang, H., Holesinger, T. G., Civale, L., Jia, Q. X., & Foltyn, S. R. (2008). Improved microstructure and enhanced low-field J_c in ($Y_{0.67}Eu_{0.3}$) $Ba_2Cu_3O_{7-\delta}$ films, 21, 1–5.