UNIVERSITI PUTRA MALAYSIA

SYNTHESIS, MORPHOLOGY AND SUPERCONDUCTING-PROPERTY CHARACTERIZATION OF Mg_xB_2 AND MgB_2 WITH SiB_4 ADDITIONS

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By

MOHD FAISAL BIN MOHD ARIS

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

October 2009

Dedications

To my wife, family and friends

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

SYNTHESIS, MORPHOLOGY AND SUPERCONDUCTING-PROPERTY CHARACTERIZATION OF Mg_xB_2 AND MgB_2 WITH SiB_4 ADDITIONS

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October 2009

Chairman: Professor Abdul Halim Bin Shaari, PhD

Faculty: Faculty of Science

This project deals with the processing of MgB_2 bulk samples and the effect of addition of SiB_4. The correlation between fabrication parameters and superconducting properties was studied. MgB_2 bulks were prepared by a pellet-in-closed-tube (PICT) and solid-state reaction route methods. MgB_2 bulk samples were prepared at different annealing temperatures of 650°C, 700°C, 750°C and 800°C. AC susceptibility measurements showed a sharp superconducting transition in all the samples. However, MgB_2, samples annealed at 650°C showed slightly low volume susceptibility compared to other samples. The critical current density (J_c) was measured using ac magnetic susceptibility measurement method. The highest critical density in magnetic field, $J_c(T,H)$ from the magnetization measurements was recorded at 3.0×10^4A/cm^2 (5K, 6T) and 7.0×10^3 A/cm^2 (20K, 4T) for the 650°C annealed MgB_2. X-ray diffraction (XRD) analysis showed small quantities of MgO and unreacted Mg phases as impurities in the samples. The second part of the work concentrates on Mg non-stoichiometry in Mg_xB_2 (x = 0.8, 1.0 and 1.2) samples. The critical temperature (T_c), was significantly reduced in sample x = 0.8 and x = 1.2 as

compared to sample with x=1.0. The $Mg_{0.8}B_2$ sample shows the highest J_c at 5K and 20K, followed by $Mg_{1.2}B_2$ and MgB_2 samples, respectively. MgB_2 shows the lowest J_c for all temperatures. The XRD results show that in the samples annealed at 650°C there is totally no unreacted Mg in the Mg-deficient sample, but some unreacted Mg exist in the nominal and Mg-excess samples. Unreacted Mg decreased with increasing annealing temperature. At 800°C, no unreacted Mg was observed in all samples. Mg-deficient cause lattice distortion on MgB_2 and create it own pinning centre base on structure defect and this will lead to enhance J_c. The final part of the work is mainly focused on SiB_4 addition on MgB_2 bulk. MgB_2 added with 0, 1, 2, 5 and 10 wt% of SiB_4 showed a sharp superconducting transition in all the samples, without significant change in critical temperature (T_c). The critical current density in applied field $J_c(H)$ of SiB_4 added samples improved significantly. $J_c(H)$ (both at 5K and 20K, T > 5T) improvement in high fields is the highest for 1.0 wt% (annealed at 650°C) and 0.5 wt% (annealed at 800°C). The value of magnetic J_c reached as high as 9.2×10^3A/cm^2 (6T, 5K). The improvement in $Jc(H)$ of the SiB_4 added samples is due to the enhanced flux pinning. X-ray diffraction (XRD) analysis showed small quantities of MgO, Mg and MgSi as impurities in the MgB_2 matrix. XRD analysis indicates that a small amount of SiB_4 was decomposed into MgSi and unreacted Mg impurity phase remained in MgB_2 matrix, and SiB_4 addition act as effective pinning centres. This work suggests that addition of SiB_4 significantly enhance $J_c(H)$ in MgB_2 superconductor.

Abstrak thesis yang dikemukakan kepada Senat Universiti Putra Malaysia bagi memenuhi keperluan Ijazah Master Sains

SINTESIS, MORFOLOGI DAN PENCIRIAN SIFAT SUPERKONDUKTOR Mg_xB_2 DAN MgB_2 DENGAN PENAMBAHAN SiB_4.

Oleh

MOHD FAISAL BIN MOHD ARIS

Oktober 2009

Pengerusi : Professor Abdul Halim Bin Shaari, PhD

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Projek ini berkaitan dengan pemprosesan sampel pukal MgB_2 dan kesan penambahan SiB_4. Pertalian di antara parameter fabrikasi dan sifat-sifat superkonduktor dikaji terperinci. MgB_2 pukal disediakan dengan teknik serbuk dalam tiub tertutup (PICT) dan kaedah tindak balas keadaan pepejal. Sampel pukal MgB_2 disediakan pada suhu 650°C, 700°C, 750°C dan 800°C. Pengukuran kerentanan arus ulang alik menunjukkan suhu genting (T_c) yang jelas untuk semua sampel, kecuali sampel yang dirawat pada 650°C menunjukkan T_c berkurang jika dibandingkan dengan sampel yang lain. Ketumpatan arus genting (J_c) telah diukur menggunakan kaedah kerentetan magnetik ulang alik. Pengukuran pemagnetan menunjukkan MgB_2 dengan rawatan haba 650°C mencapai ketumpatan arus genting angkutan dalam medan $J_c(H)$ setinggi 3.0×10^4A/cm^2 (5K, 6T) dan 7.0×10^3 A/cm^2 (20K, 4T). Analisis XRD menunjukkan kuantiti kecil MgO dan fasa Mg yang tidak bertindak balas sebagai bendasing di dalam sampel. Dalam bahagian kedua kajian adalah tertumpu kepada Mg bukan stoikiometri dalam sampel Mg_xB_2 (x=0.8, 1.0 and 1.2). T_c telah berkurang dengan nyata dalam sampel x=0.8 and x=1.2 jika dibanding dengan x=1.0. $Mg_{0.8}B_2$ menunjukkan J_c tertinggi pada 5K dan 20K, diikuti oleh $Mg_{1.2}B_2$ dan MgB_2, disebabkan bendasing yang bertindak sebagai teras pengepin itu

meningkatkan nilai J_c. Corak XRD sampel yang dirawat pada 650°C tiada Mg yang bertindak balas dalam sampel Mg-defisien, tetapi Mg yang tidak bertindak balas wujud dalam nominal dan Mg-lebihan. Mg yang tidak bertindak balas menjadi berkurang dengan penambahan suhu rawatan haba. Pada 800°C, semua Mg didapati bertindak balas sepenuhnya. Mg-defisien menyebabkan kekisi pada MgB_2 terganggu dan menghasilkan pusat pengepinannya sendiri hasil dari kecacatan struktur dan ini meningkatkan J_c. Bahagian akhir tertumpu kepada kesan penambahan SiB_4 ke atas MgB_2 pukal. MgB_2 ditambah dengan 0, 1, 2, 5 dan 10 wt% SiB_4 menunjukkan peralihan yang jelas dalam semua sampel, tanpa perubahan yang besar dalam suhu genting (T_c). Ketumpatan arus genting angkutan dalam medan $J_c(H)$ dalam sampel penambahan SiB_4 menunjukkan pertambahan yang ketara. $J_c(H)$ (pada 5K dan 20K, T > 5T) pertambahan tertinggi dalam medan tinggi ini ialah untuk 1.0 wt% (rawatan haba pada 650°C) dan 0.5 wt% (rawatan haba pada 800°C) penambahan SiB_4. Nilai ketumpatan arus genting angkutan dalam medan $J_c(H)$ mecapai setinggi 9.2×10^3A/cm^2 (6T, 5K). Peningkatan $Jc(H)$ dalam sampel yang ditambah SiB_4 disebabkan oleh peningkatan pengepinan fluks akibat daripada penambahan SiB_4. Analisis XRD menunjukkan sedikit bendasing MgO, Mg dan MgSi dalam matrik MgB_2. Analisis XRD juga menunjukkan sebahagian kecil SiB_4 terurai menjadi MgSi dan fasa bendasing Mg yang tidak bertindak balas kekal dalam matrik MgB_2, SiB_4 bertindak sebagai pusat pengepinan yang efektif. Kajian ini menunjukkan penambahan SiB_4 menyebabkan penambahan ketara J_c (H) dalam superkonduktor MgB_2.

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I certify that an Examination Committee has met on date of viva voce to conduct the final examination of Mohd Faisal Bin Mohd Aris on his Master thesis entitle “Synthesis, Morphology and Superconducting-Property Characterization of Mg_xB_2 and MgB_2 with SiB_4 additions” in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (High Degree) Regulation 1981. The Committee recommends that the student be awarded the Master of Science

Member of the Examination Committee were as follows:

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This thesis was submited 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 Commitee were as follows:

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Date: 11 February 2010

DECLARATION

I declare that the thesis is my original work except for quotations and citation 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.

MOHD FAISAL BIN MOHD ARIS

Date: 12 June 2009

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a, b, c	Lattice parameters
A	Amperes
Bi 2223	Family member in $Bi_2Sr_2Ca_nCu_{n+1}O_{6+2n}$, n=2
BCS	Bardeen-Cooper-Schrieffer
C	Celcius
Dc	Direct current
$\boldsymbol{F_p}$	Pinning force
H	Magnetic field
H_c	Critical field strength
H_{c1}	Lower Critical Field
H_{c2}	Higher Critical Field
H_{irr}	Irreversibility field
HTS	High Temperature Superconductor
J_c	Critical current density
$J_c(H)$	Critical current density in applied field
K	Kelvin
M(T)	Magnetization versus Temperature
Oe	Oersted
PICT	Powder-in-closed-tube
SEM	Scanning Electron Microscope
T	Tesla
T_c	Critical Temperature
$T_{c(0)}$, $T_{c(zero)}$	Critical Temperature Zero
V	Volume

wt%	weight percent
XRD	X-ray diffraction
YBCO	$YBa_2Cu_3O_7$
ZFC	zero-field-cooled
ξab, ξc	Coherence Length
ΔT_c	Change on critical current temperature

CHAPTER 1

INTRODUCTION

This chapter is about the history and basic properties of superconductivity such as zero dc resistance, perfect diamagnetism and flux pinning. The main research objective is discussed at the end of the chapter.

1.1 Brief history about superconductivity.

In 1911, H. Kamerlingh Onnes, who was the first to liquefy helium in 1908, began to investigate the electrical properties of metal at extremely low temperature. He found that resistivity of mercury dropped from 0.03 Ω to 3 × 10^{-6} Ω within 0.01 K temperature range after cooled below 4.2K. (Onnes, 1911).

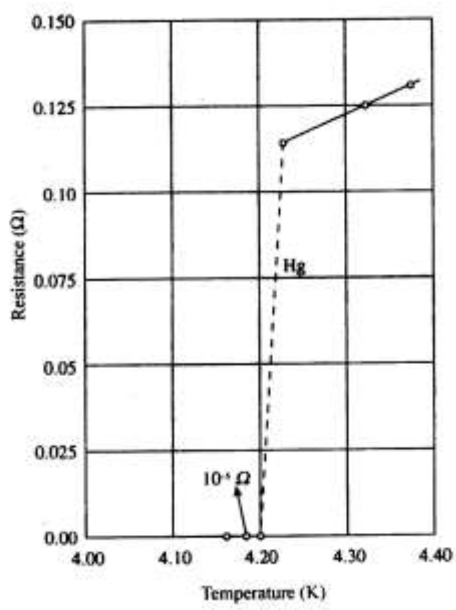

Figure 1.1: Electrical resistance of Hg at low temperature (as reported by Onnes in 1911) which showed a transition temperature at 4.2K

More elements were discovered as superconductors later such as tin, lead, indium, aluminium, niobium and several alloys which superconduct at very low temperatures and known as conventional superconductors. Among conventional superconductors, MgB_2 has the highest transition temperature (39K). Onnes also noticed that superconductivity was influenced by external magnetic field which can drive a superconductor back to normal state.

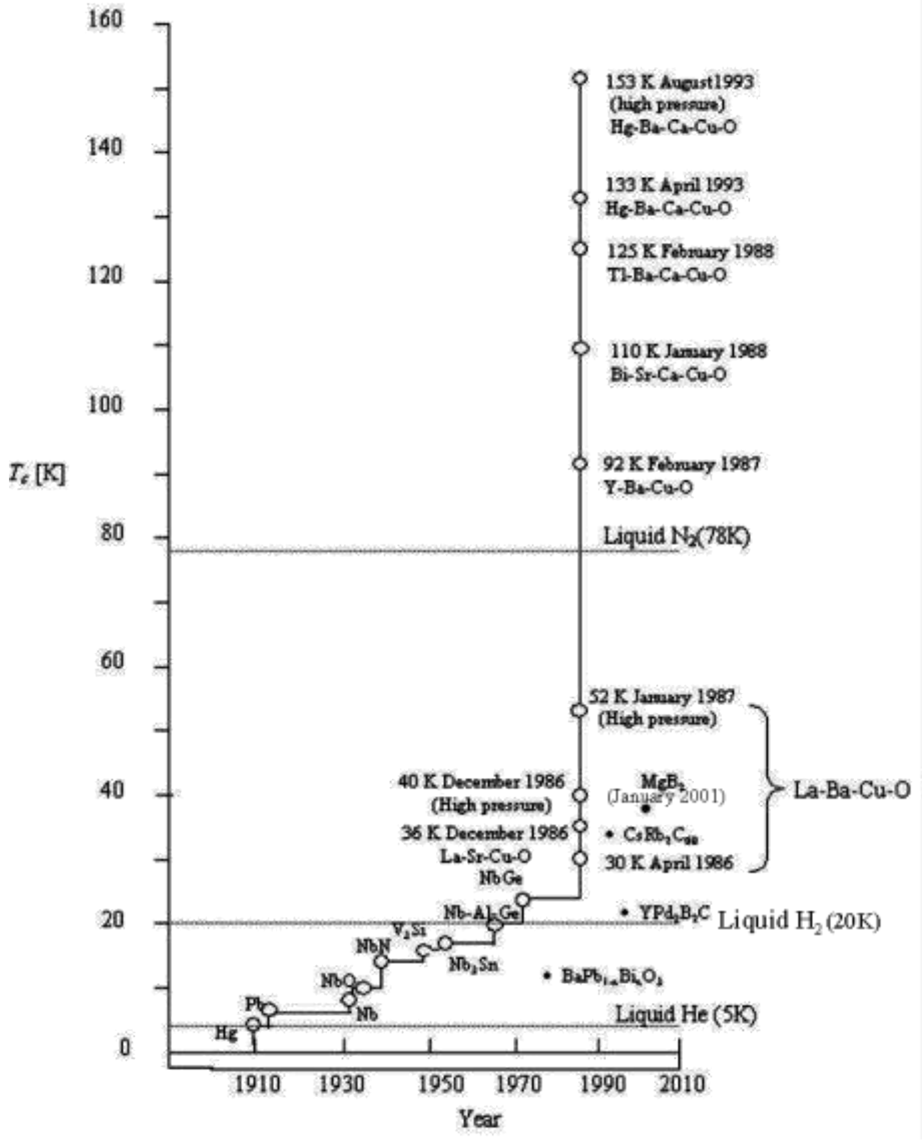

Figure 1.2: The evolution of T_c according to the year of discovery (Zhou, 2004)

1.2 Basic properties of superconductor

Superconductivity is characterized by two important properties which are zero dc resistivity and perfect diamagnetism.

1.2.1 Zero dc resistivity

The zero dc resistivity occurred in superconductor because at low temperature the the charge carriers known as Cooper pairs are able to move freely without any resistance. The temperature at which the superconductor losses resistance is called superconducting transition temperature or critical temperature (T_c) where resistivity sharply drops to zero for direct current (dc) and nearly all resistance to the flow of alternating current. The microscopic mechanism of superconductivity in metal and alloys can be explained by the Bardeen-Cooper-Schrieffer (BCS) theory.

1.2.2 Perfect diamagnetism

Twenty two years after the discovery of zero resistivity by Onnes in 1911, Walther Meissner and Robert Ochsenfeld found that when a superconductor is cooled below its critical temperature, perfect diamagnetism occurred when magnetic field is applied. This is later known as the Meissner effect. Perfect diamagnetism occurs when surface current induced by a superconductor expels the externally applied field. The limit of external magnetic field strength at which a superconductor can exclude the field is known as the critical field, (H_c). Figure 1.33 shows the effect of magnetic field on a (a) perfect diamagnet (b) normal diamagnet and (c) paramagnetic

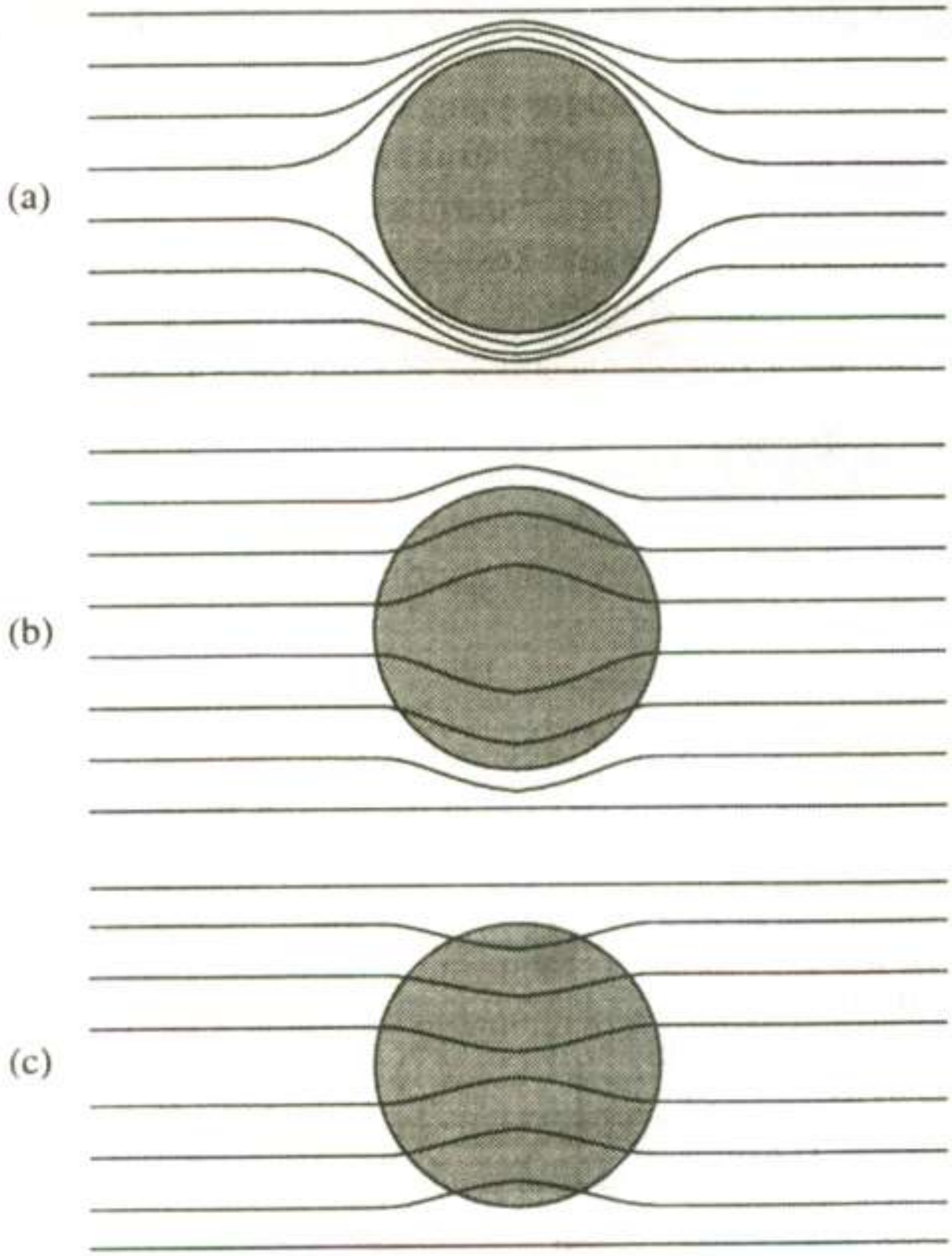

Figure 1.3: Flux density in a (a) perfect diamagnet such as superconductor, (b) normal diamagnet and (c) paramagnet (Abd-Shukor, 2004)