



UNIVERSITI PUTRA MALAYSIA

**SUPERCONDUCTING PROPERTIES OF MgB_2 AFTER REACTION WITH
SILICON AND CARBON-CONTAINING ADDITIVES**

TAN KWEE YONG

FS 2011 71

**SUPERCONDUCTING PROPERTIES OF MgB₂ AFTER REACTION WITH SILICON AND
CARBON-CONTAINING ADDITIVES**

TAN KWEE YONG

**Thesis Submitted to the School of Graduate Studies, Universiti PutraMalaysia, in Fulfilment of the
Requirements for the degree of Master of Science**

November 2011

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

**SUPERCONDUCTING PROPERTIES OF MgB₂ AFTER REACTION WITH
SILICON AND CARBON-CONTAINING ADDITIVES**

By

TAN KWEE YONG

November 2011

Chair: Chen Soo Kien, PhD

Faculty: Faculty of Science

SiC is one of the promising dopants that effectively improves the critical current density (J_c) of MgB₂ by substituting C into B-site and enhances electron scattering. However, the roles of Si and C in influencing the superconducting properties of MgB₂ are not fully understood. Furthermore, systematic study on the optimum dopant addition level and effect of sintering temperature are required in order to provide further insight into how SiC or both Si and C enhance J_c . In this study, nano-SiC and combination of nano-Si and nano-C (Si+C) that made of similar ratio to that of SiC [up to 15 weight percentage (wt.%)] was reacted with Mg+B powder by *in situ* solid state method. These bulks were sintered at 650°C and 850°C, respectively. Characterizations are performed by using X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), and Magnetic Property Measurement System (MPMS). These samples were compared in terms of phase formation, lattice properties, microstructure and superconducting properties.

At 650°C, samples reacted with SiC show smaller a -axis and more vigorous lattice distortion because of higher C substitution at B site as compared to same amount of

(Si+C) addition. This is due to the reactive form of C atoms released from Mg-SiC

reaction with lower Gibbs free energy. Higher C substitution in SiC reacted sample results in more severe degradation in superconducting transition temperature (T_c) arising from more severe lattice distortion. Samples reacted with SiC (up to 5 wt.%) show stronger improvement in J_c at both 5 K and 20 K mainly because of smaller grains that enhance grain boundary pinning and degraded crystallinity due to lattice defect.

At 850°C, (Si+C) reacted samples have greater extent of a -axis contraction and more severe lattice distortion because of higher C substitution than those samples reacted with SiC. Such phenomenon is probably due to the availability of more C in which separate Si and C particles are used or effect of higher sintering temperature. Higher level of C substitution in (Si+C) reacted samples leads to more severe T_c suppression because of more severe lattice distortion. On the other hand, (Si+C) reacted samples show stronger J_c improvement at both 5 K and 20 K due to higher C substitution at B-site that further enhances electron scattering.

As a conclusion, for sintering at 650°C, reaction of SiC with MgB₂ is preferred while sintering at 850°C, reaction of (Si+C) with MgB₂ is favored as higher C substitution and more lattice defects occur in these samples which effectively enhance the J_c . Among all samples, the J_c of 5 wt.% SiC reacted MgB₂ at 650°C, as compared to the pure sample, is improved by a factor of two at 5 K for the applied field of 6 T and 60 % more at 20 K for the applied field of 4 T, respectively. This improvement is due to degraded crystallinity and higher C substitution that further scatter the electrons and smaller grains that enhance grain boundary pinning.

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

**SIFAT-SIFAT KESUPERKONDUKTORAN MgB₂ SELEPAS BERTINDAK
BALAS DENGAN BAHAN TAMBAHAN MENGANDUNGI SILIKON DAN
KARBON**

Oleh

TAN KWEE YONG

November 2011

Pengerusi: Chen Soo Kien, PhD

Fakulti: Fakulti Sains

SiC adalah salah satu dopan yang dapat meningkatkan ketumpatan arus genting (J_c) MgB₂ secara berkesan dengan menggantikan C ke dalam tapak B dan juga meningkatkan penyebaran elektron. Tetapi, fungsi Si dan C dalam mempengaruhi sifat-sifat kesuperkonduktoran MgB₂ belum difahami sepenuhnya. Tambahan pula, kajian yang sistematik terhadap penambahan dopan optimum dan kesan suhu pemanasan adalah diperlukan agar memberi pandangan mendalam terhadap bagaimana SiC atau kedua-dua Si dan C dapat meningkatkan J_c . Dalam kajian ini, nano-SiC atau kombinasi nano-Si dan nano-C berasingan (Si+C) yang dicampur pada nisbah yang sama kepada SiC [sehingga 15 peratus keberatan (wt.%)] telah ditindak-balas dengan serbuk Mg+B secara kaedah keadaan pepejal *in situ*. Sampel-sampel ini dipanaskan pada 650°C and 850°C secara berasingan. Pencirian sampel dijalankan dengan pembelauan sinar X (XRD), mikroskopi imbasan elektron (SEM) dan sistem penyukuran sifat magnetik (MPMS). Sampel-sampel ini telah dibandingkan dari segi pembentukan fasa, sifat kekisi, mikrostruktur dan sifat kesuperkonduktoran.

Pada 650°C, sampel yang bertindak balas dengan SiC menunjukkan paksi- a yang lebih

kecil, struktur kekisi yang lebih herot disebabkan penggantian C ke dalam B yang lebih tinggi ke dalam tapak B jika dibandingkan dengan penambahan (Si+C) yang sama tahap.

Ini adalah kerana C atom yang lebih reaktif dibebaskan dari tindak-balas Mg-SiC dengan tenaga bebas Gibbs yang lebih rendah. Penggantian C yang tinggi dalam sampel juga memberikan kesan penurunan suhu genting (T_c) yang lebih serius oleh kerana herotan kekisi yang nyata. Sampel yang bertindak balas dengan SiC (sehingga 5 wt.%) menunjukkan peningkatan J_c yang lebih baik pada 5 K dan 20 K disebabkan lebih banyak butiran kecil yang meningkatkan pengepitan sempadan butiran dan kehabluran yang ternyahgred akibat kecacatan kekisi.

Pada 850°C, sampel yang bertindak balas dengan (Si+C) mempunyai susutan paksi- a yang lebih teruk dan kekisi yang lebih herot disebabkan penggantian C ke dalam B yang lebih banyak dibandingkan dengan sampel yang bertindak-balas dengan SiC. Ini adalah kerana terdapatnya lebih banyak C dengan menggunakan Si dan C yang berasingan dan penggantian yang digalakkan oleh suhu pemanasan yang lebih tinggi. Penggantian C yang lebih tinggi dalam sampel yang bertindak balas dengan (Si+C) turut menyebabkan penurunan T_c yang lebih nyata kerana kekisinya lebih herot. Akan tetapi, peningkatan J_c pada 5 K dan 20 K telah ditunjukkan dalam sampel yang bertindak balas dengan (Si+C) akibat penggantian C yang lebih banyak untuk penyebaran elektron.

Secara kesimpulan, bagi pemanasan pada 650°C, MgB₂ adalah digalakkan bertindak balas dengan SiC untuk mendapat J_c yang lebih tinggi manakala untuk pemanasan pada 850°C, MgB₂ adalah digalakkan bertindak balas dengan (Si+C) kerana penggantian C yang lebih banyak untuk menyebarkan elektron dan kecacatan kekisi yang lebih nyata

ditunjukkan dalam sampel-sampel tersebut. Sekiranya dibandingkan dengan sampel tulen, peningkatan J_c yang ditunjukkan dalam sampel yang bertindak balas dengan 5 wt.% SiC pada 650°C , adalah sebanyak dua kali ganda pada 5 K, 6 T dan 60 % lebih banyak pada 20 K, 4 T dalam keadaan suhu dan medan magnetik berlainan. Ini adalah disebabkan kehabluran yang lebih nyahged dan penggantian C yang lebih banyak untuk menyebarkan lagi elektron serta butiran lebih kecil yang meningkatkan lagi pengepunan sempadan butiran.



ACKNOWLEDGEMENTS

I would firstly like to express my deepest gratitude to my supervisor, Dr. Chen Soo Kien for his unwavering encouragement, guidance which enabled me to complete this study. I appreciate this great opportunity and platform which enhanced me towards better understanding of a research in related to materials with improved superconductivity.

Secondly, it is my pleasure to learn from my co-supervisors: Prof. Dr. Abdul Halim Shaari and Dr. Tan Kar Ban. They had been giving me constant advices and supports in order to improve the quality of this research.

I would like to extend my appreciations to my lab colleagues, Ms. Ng Siau Wei, Mr. Wong Jen Kuen, Ms. Tan Kim Lee and others for their help towards the success of this research. Also, I thank Material Science and Metallurgy Department, University of Cambridge, Dr. Lim Kean Pah, En. Zain, Pn. Kamsiah and Pn. Zahidah for their assistance in measurements and analyses.

Last but not the least, I offer my regards and blessings to all of those who supported me in any respect during the completion of the project especially my family and friends who have been showering me with their encouragement and concern.

The financial support from Ministry of Science, Technology and Innovation, Malaysia (MOSTI) via ScienceFund (03-01-04-SF0920) is also gratefully appreciated.

I certify that an Examination Committee has met on 2nd November 2011 to conduct the final examination of Tan Kwee Yong on his thesis entitled “Superconducting properties of MgB₂ Reacted with Silicon and Carbon-Containing Additives” in accordance with 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 Examination Committee were as follows:

Hishamuddin Zainuddin, PhD

Associate Professor
Department of Physics
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Wan Mahmood Mat Yunus, PhD

Professor
Department of Physics
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Jumiah Hassan, PhD

Associate Professor
Department of Physics
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Ahmad Kamal Yahya, PhD

Professor
School of Physics and Materials Studies
Faculty of Applied Science
Universiti Teknologi Mara
Malaysia
(External Examiner)

SEOW HENG FONG, PhD

Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia
Date: 20 December 2011

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:

Chen Soo Kien, PhD

Faculty of Science
Universiti Putra Malaysia
(Chairman)

Abdul Halim Shaari, PhD

Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

Tan Kar Ban, PhD

Faculty of Science
Universiti Putra Malaysia
(Member)

BUJANG BIN KIM HUAT, PhD

Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia
Date

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.

TAN KWEE YONG
Date: 2nd November 2011



TABLE OF CONTENTS

	Page
ABSTRACT	iii
ABSTRAK	v
ACKNOWLEDGEMENTS	viii
APPROVAL	ix
DECLARATION	xi
LIST OF TABLES	xii
LIST OF FIGURES	xiii
LIST OF ABBREVIATIONS	xviii
CHAPTER	
1 INTRODUCTION	1
1.1 Background of the Project	1
1.2 Motivation and Objectives	2
1.3 Markets and Potentials	4
1.4 Applications of Superconductors	6
1.4.1 Large Scale Applications	8
1.4.2 Superconducting Electronics Applications	10
2 BACKGROUND AND BASIS OF SUPERCONDUCTOR	12
2.1 A Brief History of Superconductivity	12
2.2 Basic properties of Superconductor	14
2.2.1 Zero Resistance	14
2.2.2 The Meissner-Ochsenfeld Effect	15
2.3 Superconducting Materials	17
2.4 The Theory of Superconductivity (BCS Theory)	18
2.5 Classification of Superconductor with Ginzburg-Landau Coefficient, κ	19
2.5.1 Type I Superconductors	21
2.5.2 Type II Superconductors	22
2.6 Destruction of Superconductivity by Magnetic Field	23
2.7 Mixed State in Type II Superconductors	25
2.8 Vortex Motion, Vortex Pinning and Critical Current Density	26
3 LITERATURE REVIEW	29
3.1 Introduction	29
3.1.1. Discovery of MgB ₂ Superconductor	29
3.2 Some Basic Properties of MgB ₂	29
3.2.1 Crystal Structure	29
3.2.2 Transparency of Grain Boundary to Current (Free of Weak Links)	30
3.2.3 Anisotropy of MgB ₂	31
3.3 Comparison of Cost	31
3.4 Sample Preparation and Phase Formation of MgB ₂	32
3.4.1 Phase Diagram of Mg-B	33

3.4.2 Sintering Environment of MgB ₂	34
3.4.3 Sintering Temperature and Sintering Duration	35
3.4.4 <i>In situ/Ex situ</i> Sample Preparation Method	37
3.4.5 Effect of Particle Size and Purity of Raw Materials	37
3.5 Morphology and Microstructure of MgB ₂	38
3.6 Chemical Doping in MgB ₂	41
3.6.1 SiC Doping	41
3.6.2 Si Doping	46
3.6.3 C Doping	47
4 EXPERIMENTAL METRHODS AND MEASUREMENTS	52
4.1 Sample Preparation	52
4.1.1 Overall Workflow of the Study	53
4.2 Raw Materials	54
4.3 Characterizations	56
4.3.1 X-ray Diffraction (XRD)	56
4.3.2 Scanning Electron Microscopy (SEM)	58
4.3.3 Superconducting Quantum Interference Device (SQUID)	60
5 RESULTS AND DISCUSSION	64
5.1 MgB ₂ Reacted with SiC	64
5.1.1 Sintering at 650°C	64
5.1.2 Sintering at 850°C	77
5.1.3 Summary of Superconducting Properties of the Pure and SiC Added MgB ₂ Reacted at 650°C and 850°C	89
5.2 MgB ₂ Reacted with (Si+C)	94
5.2.1 Sintering at 650°C	95
5.2.2 Sintering at 850°C	105
5.2.3 Summary of Superconducting Properties of the Pure and (Si+C) Added MgB ₂ Reacted at 650°C and 850°C	117
5.3 Comparisons of Superconducting Properties between SiC and (Si+C) Addition at 650°C	123
5.3.1 Superconducting Transition Temperature	123
5.3.2 Critical Current Density	124
5.4 Comparisons of Superconducting Properties between SiC and (Si+C) Addition at 850°C	128
5.4.1 Superconducting Transition Temperature	128
5.4.2 Critical Current Density	129
6 CONCLUSIONS AND FUTUREWORK	134
REFERENCES	137
BIODATA OF THE STUDENT	158
LIST OF PUBLICATIONS	159