

การปรับปรุงสภาพนำความร้อนของอะลูมินาเพื่อใช้ทำเพลเทียร์อิไลเมนต์

นางสาวบงกช เปี่ยมเพิ่มพูน



วิทยานิพนธ์นี้เป็นส่วนหนึ่งของการศึกษาตามหลักสูตรปริญญาวิทยาศาสตรมหาบัณฑิต

สาขาวิชาเทคโนโลยีเซรามิก ภาควิชาวัสดุศาสตร์

คณะวิทยาศาสตร์ จุฬาลงกรณ์มหาวิทยาลัย

ปีการศึกษา 2545

ISBN 974-17-0850-5

ลิขสิทธิ์ของจุฬาลงกรณ์มหาวิทยาลัย

IMPROVEMENT OF THERMAL CONDUCTIVITY OF ALUMINA FOR PELTIER ELEMENT

Miss Bongkoch Piempempon

A Thesis Submitted in Partial Fulfillment of the Requirements
for the Degree of Master of Science in Ceramic Technology

Department of Material Science

Faculty of Science

Chulalongkorn University

Academic Year 2002


ISBN 974-17-0850-5


Thesis Title Improvement of thermal conductivity of alumina for Peltier element
By Miss. Bongkoch Piempermpoon
Department Material science
Thesis Advisor Chair Professor Shigetaka Wada, Ph.D.
Thesis Co-advisor Sirithan Jiemsirilers, Ph.D.

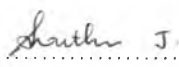
Accepted by the Faculty of science, Chulalongkorn University in Partial
Fulfillment of the Requirements for the Master 's Degree


 Dean of Faculty of science
(Associate Professor Wanchai Phothiphichitr, Ph.D.)

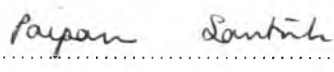
THESIS COMMITTEE

 Chairman
(Associate Professor Saowaroj Chuayjuljit)

 Thesis Advisor
(Chair Professor Shigetaka Wada, Ph.D.)

 Thesis Co-advisor
(Sirithan Jiemsirilers, Ph.D.)

 Member
(Associate Professor Supatra Jinawath, Ph.D.)


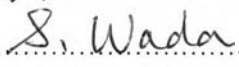
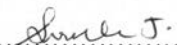
 Member
(Associate Professor Paipan Santisuk)

บงกช เปี่ยมเพิ่มพูน : การปรับปรุงสภาพนำความร้อนของอะลูมินาเพื่อใช้ทำเพลเทียร์อีลีเมนต์. (IMPROVEMENT OF THERMAL CONDUCTIVITY OF ALUMINA FOR PELTIER ELEMENT) อ.ที่ปรึกษา : ศ.ดร. ชิกตากะ วาดะ, อ.ที่ปรึกษาร่วม : อ.ดร. ศิริธันว์ เจียมศิริเลิศ, 139 หน้า. ISBN 974-17-0850-5.

งานวิจัยนี้ได้ศึกษาผลของผงอะลูมินา สารเจือ และอุณหภูมิการขึ้นเทอร์โม ที่มีต่อสมบัตินำความร้อนของแผ่นรองอะลูมินาสำหรับใช้ทำเพลเทียร์อีลีเมนต์ ซึ่งแผ่นรองอะลูมินานี้ควรมีความแข็งแรงเชิงกลสูงและมีราคาถูก ในงานวิจัย ผงอะลูมินาที่ใช้คือ AKP-30 และ AES-11 ซึ่งมีความบริสุทธิ์มากกว่า 99.99 และ 99.8 เปอร์เซ็นต์ตามลำดับ ในขณะที่ สารเจือที่ใช้คือ แมกนีเซียมออกไซด์ และ เซอร์โคเนียมออกไซด์ เตรียมผงอะลูมินาเติมด้วยสารเจือ 0.5 1.5 3.0 7.5 เปอร์เซ็นต์โดยน้ำหนัก และเผาขึ้นเทอร์โมในช่วง 1450 – 1650 องศาเซลเซียส เป็นเวลา 2 ชั่วโมง.

AKP-30 และ AES-11 ที่มี 0.5 เปอร์เซ็นต์โดยน้ำหนักของแมกนีเซียมออกไซด์ และ 1.5 3.0 7.5 เปอร์เซ็นต์โดยน้ำหนักของเซอร์โคเนียมออกไซด์ สามารถขึ้นเทอร์โมให้มีความหนาแน่นใกล้เคียงกับค่าความหนาแน่นในทางทฤษฎี การนำความร้อนของ AKP-30 มีค่าอยู่ในช่วง 30 – 38 วัตต์ต่อเมตรเคลวิน ในขณะที่การนำความร้อนของ AES-11 มีค่าอยู่ในช่วง 27 – 34 วัตต์ต่อเมตรเคลวิน ซึ่งพบว่า AKP-30 มีค่าการนำความร้อนที่ดีกว่า AES-11 เพียงเล็กน้อย, AES-11 ที่ได้ทำการขึ้นเทอร์โมที่ 1550 – 1650 องศาเซลเซียส มีค่าความแข็งแรงเชิงกลมากกว่า 400 เมกะปาสคาล ซึ่งให้ค่าความแข็งแรงเชิงกลและการนำความร้อนที่เป็นที่น่าพอใจในงานวิจัยนี้

ภาควิชาวัสดุศาสตร์
สาขาวิชา เทคโนโลยีเซรามิก
ปีการศึกษา 2545

ลายมือชื่อนิสิต.....
ลายมือชื่ออาจารย์ที่ปรึกษา..... S. Wada
ลายมือชื่ออาจารย์ที่ปรึกษาร่วม.....

4372310823 : MAJOR CERAMIC TECHNOLOGY

KEY WORD: PELTIER ELEMENT / THERMAL CONDUCTIVITY / MECHANICAL
STRENGTH / DENSITY


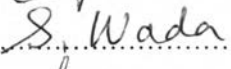

BONGKOCH PIEMPERMPOON : IMPROVEMENT OF THERMAL CONDUCTIVITY OF
ALUMINA FOR PELTIER ELEMENT.

THESIS ADVISOR : PROF. SHIGETAKA WADA, Ph.D., THESIS CO-ADVISOR : SILITAN
JIEMSILILERD, Ph.D. 139 pp. ISBN 974-17-0850-5.

This study concentrated on improvement of thermal conductivity of alumina substrate for Peltier element as functions of alumina powders, dopants, and sintering temperatures. Alumina substrate also should have high mechanical strength and cheap cost. In this study, AKP-30 and AES-11 were used as alumina powder while MgO and ZrO₂ were used as dopants. Alumina powders were doped with 0.5 wt% of MgO and 1.5, 3.0 and 7.5 wt% of ZrO₂ and were prepared by a conventional oxide mixing technique and sintered at temperature ranging from 1450 – 1650 °C for 2 hours.

AKP-30 and AES-11 with 0.5 wt% of MgO and 1.5, 3.0, 7.5 wt% of ZrO₂ dopants could be sintered to nearly theoretical density. Thermal conductivity of AKP-30 was in the range of 30 – 38 W/m·K while that of AES-11 was in the range of 27 - 34 W/m·K. Therefore AKP-30 had a better thermal conductivity than AES-11, but not so significantly. The mechanical strength of most of AES-11 specimen sintered at 1550 – 1650 °C was higher than 400 MPa. Apparently the mechanical strength and thermal conductivity measured in this research had met the targeted values.

Department Material Science
Field of study Ceramic Technology
Academic year 2002

Student's signature.....
Advisor's signature.....
Co-advisor's signature.....



Acknowledgements

I would like to express to my deep gratitude to my advisor, Professor Dr. Shigetaka Wada, for his encouragement, consistent guidance and for all that I have learnt from his throughout this research. His advises never failed to inspire the good idea and increase my motivation. I would like to extend my gratitude to my co-advisor, Dr. Sirithan Jiemsirilers, who gave me a good advice.

I would like to acknowledge Mr. Tanakorn Wasanapienpong for his advice. My works would not be completed without his helpful and invaluable suggestions.

I would like to thank the National Metal and Materials Technology Center (MTEC) and graduate school of Chulalongkorn University especially the AISIN, Japanese company for the research financial support.

My thanks are also extended to Mr. Panyapong Kimphan for his helpful and support.

Thanks to all my friends at the Department of Materials Science for their friendship and supports.

Finally, I would like to express my gratitude to my family for their loves, understanding and encouragement.

CONTENTS

	Page
Abstract (Thai).....	iv
Abstract (English).....	v
Acknowledgements.....	vi
Contents.....	vii
Lists of Tables.....	x
Lists of Figures.....	xi
Chapter 1 Introduction.....	1
Chapter 2 Literature Review.....	4
2.1 Alumina.....	4
2.2 Sintering of alumina.....	7
2.3 Influence of additives.....	9
2.3.1 Effect of MgO.....	9
2.3.2 Effect of ZrO ₂	11
2.3.3 Effect of SiO ₂ , SiO ₂ -CaO.....	12
2.4 Thermal conductivity of ceramic materials.....	13
2.4.1 Thermal conduction mechanism.....	13
2.4.2 Thermal conductivity of Al ₂ O ₃	16
Chapter 3 Experimental procedures.....	18
3.1 Raw materials and characterizations	18
3.1.1 Starting raw materials.....	18
3.1.2 Raw material characterizations.....	19
3.1.2.1 Particle size distribution determination.....	19
3.1.2.2 Microstructure examination by SEM.....	19
3.2 Composition and preparation of alumina powder.....	20
3.3 Characterization of sintered specimens.....	23
3.3.1 Density measurement.....	23
3.3.1.1 Specific gravity.....	23
3.3.1.2 Theoretical density.....	23

CONTENTS (Cont.)

	Page
3.3.1.3 Relative density.....	24
3.3.2 Water absorption measurement.....	24
3.3.3 Microstructure examination by SEM.....	25
3.3.4 Strength measurement.....	25
3.3.5 Thermal conductivity measurement.....	27
Chapter 4 Experimental results.....	28
4.1 Raw materials characterizations.....	28
4.1.1 Particle size distribution.....	28
4.1.2 Morphology of alumina powder.....	30
4.2 Preliminary sintering of AES-11 and AKP-30.....	31
4.3 Thermal conductivity and microstructure of AES-11 and AKP-30.....	32
4.4 Mechanical properties of AES-11.....	38
4.5 The effects of sintering time of AES-11.....	40
4.6 Sintering of mixed additives at 1450-1650 °C.....	42
Chapter 5 Discussion.....	43
5.1 The effects of the difference of Al ₂ O ₃ powder for density and microstructure.....	43
5.2 The effects of the difference of additives for mechanical strength.....	44
5.3 The causes of deviation in relative density.....	45
5.4 Mechanical strength.....	49
5.5 Thermal conductivity.....	53
Chapter 6 Conclusions.....	59
Chapter 7 Future work.....	60
References.....	61
Appendices.....	64
1 Classification of ceramics by function.....	65
2 Thermal conductivity of alumina from several manufacturers.....	66
3 Drawing of biaxial-flexure strength-test equipment.....	68

CONTENTS (Cont.)

	Page
4 Specific gravity, water absorption and relative density of AKP-30 and AES-11 sintered at 1450 – 1650 °C.....	71
5 The relationship between relative density and sintering temperature of AKP-30 and AES-11.....	74
6 Specific gravity, water absorption and relative density of AKP-30 and AES-11 sintered at 1650 °C.....	75
7 SEM micrograph of doped and pure of AKP-30 and AES-11.....	77
8 Specific gravity, water absorption and relative density of AES-11 sintered at 1450 – 1650 °C.....	82
9 Thickness and radius of AES-11 sintered at 1450 – 1650 °C.....	87
10 Strength measurement of AES-11 specimens sintered at 1450 – 1650 °C...92	92
11 Fracture of doped and pure AES-11 sintered at 1450 - 1650 °C	97
12 Surface roughness of AES-11 specimens sintered at 1450 – 1650 °C.....	102
13 Specific gravity, water absorption and relative density of AES-11 sintered at 1450 – 1650 °C for 1, 4, and 6 h.....	107
14 The relationship between sintering time, relative density and sintering temperature of doped and pure AES-11.....	122
15 Specific gravity, water absorption and relative density of both additives doped AKP-30 and AES-11 at 1450 – 1650 °C.....	125
16 The relationship between sintering temperature and relative density of both additives doped AKP-30 and AES-11.....	133
17 Specific gravity, water absorption and relative density of second Measurement of AKP-30 and AES-11 sintered at 1650 °C.....	135
18 Comparison of relative density of AES-11 between first and second measurement.....	137
Biography.....	139

LISTS OF TABLES

	Page
Table 2.1 Typical properties of alumina.....	5
Table 2.2 Values of intrinsic thermal conductivity of nonmetallic materials.....	14
Table 2.3 Approximate values for thermal conductivities of selected ceramic materials.....	15
Table 3.1 Oxide powders and chemicals used in experiment.....	18
Table 3.2 Compositions of alumina for Peltier element.....	21
Table 3.3 Calculated 100% density of each composition.....	24
Table 4.1 Thermal conductivity of doped and pure AKP-30 and AES-11 alumina specimens sintered at 1650 °C for 2 h.....	33
Table 4.2 The average grain size of alumina sintered at 1650 °C for 2 h.....	37
Table 5.1 Measured thermal conductivity and calculated thermal conductivity.....	54

LISTS OF FIGURES

	Page
Fig.1.1 Peltier element.....	1
Fig.1.2 Structure of Peltier element.....	2
Fig.2.1 Packing of Al and O ions in the basal plane.....	4
Fig.2.2 Schematic of the geometry of sintering of two spheres, showing six alternate paths for matter transport.....	7
Fig.2.3 Effects of MgO content on the density of sintered alumina.....	9
Fig.2.4 Thermal conductivity of alumina as a function of temperature.....	16
Fig.2.5 Purity dependence of thermal conductivity on alumina content.....	17
Fig.3.1 Flow diagram of sample preparation of Peltier element by a conventional oxide mixing process.....	22
Fig.3.2 Biaxial- flexure strength-test fixture in ASTM F394-78.....	26
Fig.3.3 Flash diffusivity apparatus (Schematic).....	27
Fig.4.1 Particle size distribution of AKP-30 and AES-11 alumina powders.....	29
Fig.4.2 (a) Microstructure of pure AKP-30 alumina powder.....	30
(b) Microstructure of pure AES-11 alumina powder.....	30
Fig.4.3 (a) The relationship between relative density and sintering temperature of AKP-30.....	31
(a) The relationship between relative density and sintering temperature of AES-11.....	32
Fig.4.4 The relationship between relative density and thermal conductivity of AKP-30 and AES-11.....	34
Fig.4.5 The relationship between the amount of ZrO ₂ and thermal conductivity of AKP-30 and AES-11.....	34
Fig.4.6 (a) SEM micrograph of 0% ZrO ₂ doped AKP-30.....	36
(b) SEM micrograph of 0% ZrO ₂ doped AES-11.....	36
Fig.4.7 The relationship between the amount of ZrO ₂ and the average grain size of AKP-30 and AES-11.....	37
Fig.4.8 The relationship between strength and sintering temperature of AES-11.....	39

LISTS OF FIGURES (Cont.)

	Page
Fig.4.9 The relationship between the relative density and sintering temperature of AES-11.....	39
Fig.4.10 The relationship between water absorption and sintering temperature of AES-11.....	40
Fig.4.11 The relationship between sintering time, relative density and sintering Temperature of AES-11 doped with 7.5 wt% ZrO ₂	41
Fig.5.1 The relationship between sintering temperature and maximum difference (%).....	46
Fig.5.2 (a) The relationship of relative density between the repeated measurement for specimens No.1, 1450 – 1650 °C.....	48
(a) The relationship of relative density between the repeated measurement for specimens No.2, 1450 – 1650 °C.....	48
Fig.5.3 SEM micrograph of 0.5 wt% MgO doped AES-11 alumina sintered at 1600 °C 2 h.....	50
Fig.5.4 SEM micrograph of 1.5 wt% ZrO ₂ doped AES-11 alumina sintered at 1600 °C 2 h.....	50
Fig.5.5 (a) Surface roughness of alumina of AISIN No.1	51
(b) Surface roughness of alumina of AISIN No.2.....	52
Fig.5.6 The relationship between relative density and thermal conductivity of AKP-30 and AES-11.....	56
Fig.5.7 The relationship between the amount of and thermal conductivity of AKP-30 and AES-11.....	56
Fig.5.8 The relationship between K_m and measured thermal conductivity(w/m•k).....	57
Fig.5.9 SEM micrograph of AISIN substrate.....	58
Fig.5.10 SEM micrograph of Y-A995 specimen sintered at 1650°C 2h.....	58