# SYNTHESIS AND CHARACTERIZATION OF METAL TELLURIDES AND ANTIMONIDES FOR USING AS THERMOELECTRIC MATERIALS

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DOCTOR OF PHILOSOPHY
IN MATERIALS SCIENCE

THE GRADUATE SCHOOL
CHIANG MAI UNIVERSITY
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A THESIS SUBMITTED TO THE GRADUATE SCHOOL IN
PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF
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THIS THESIS HAS BEEN APPROVED
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## ABSTRACT



In this research, thermoelectric materials:ZnTe and $Sb_2Te_3$were successfully synthesized by microwave heating and microwave generating of plasma for solid-state reaction.Cubic ZnTenanocrystals were synthesized from 1:1, 1.5:1 and 1.8:1 molar ratios of Zn:Te by a 900 W microwave plasma. Their green emissions were detected at 562 nm (2.21 eV) using luminescence spectrophotometry.$Sb_2Te_3$ with a rhombohedral crystal system was successfully synthesized by an environmentally benign process with a short reaction time using a 900 W irradiated microwave plasma. At 2:2, 2:1.75, and 2:1.5 molar ratios of Sb:Te, with time lengths of 10 and 20 min,

the products were pure $Sb_2Te_3$ phase with no detection of any residues. The direct energy gaps were determined to be 0.340–0.515 eV.

In addition, $Ni_3GaSb$ and $Ni_3InSb$ were successfully synthesized by alloying synthesis at high temperature in closed silica ampoules.$Ni_3GaSb$ and $Ni_3InSb$ compounds indicated metal-like characteristics in $\alpha$ and $\rho$. The power factor ($\alpha^2\rho^{-1}$) values increased with temperature and reached maximum at 1073 K. The thermal conductivity ($\kappa$) and dimensionless figure of merit $ZT$ of both sampleswere increased with temperature as well. The maximum values of the $ZT$for $Ni_3GaSb$ and $Ni_3InSb$ at 1073 Kwere achieved to be 0.022 and 0.023, respectively.

**ชื่อเรื่องวิทยานิพนธ์** การสังเคราะห์และการหาลักษณะเฉพาะของโลหะเทลลูไลด์และแอนติโมไนด์สำหรับใช้เป็นวัสดุเทอร์โมอิเล็กตริก

**ผู้เขียน** นายธวัช สุริวงษ์

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## บทคัดย่อ



งานวิจัยนี้ เป็นการสังเคราะห์วัสดุเทอร์โมอิเล็กตริก คือ ZnTeและ $Sb_2Te_3$ด้วยวิธีพลาสมาของรังสีไมโครเวฟเพื่อให้เกิดปฏิกิริยาของของแข็ง ซึ่งประสบความสำเร็จในการสังเคราะห์ZnTeที่มีขนาดผลึกในระดับนาโนเมตร ด้วยอัตราส่วนโดยโมลของ Zn:Te เท่ากับ 1:1, 1.5:1 และ 1.8:1 ที่ 900 W ไมโครเวฟ โดยสาร ZnTeนี้เรืองแสงสีเขียวที่มีความยาวคลื่น 562 nm(2.21 eV) สำหรับการสังเคราะห์สาร$Sb_2Te_3$มีการใช้ 900 Wไมโครเวฟ ที่อัตราส่วนโดยโมลของ Sb:Teเท่ากับ 2:2, 2:1.75 และ 2:1.5 และเวลาในการทำปฏิกิริยาเป็น10 และ 20 นาที ตามสภาวะการทดลองนี้ทำให้สังเคราะห์สาร $Sb_2Te_3$มีความบริสุทธิ์เมื่อทำการวิเคราะห์หาช่องว่างพลังงานพบว่าอยู่ในช่วง 0.340-0.515 eV

นอกจากนี้ยังได้สังเคราะห์สารประกอบ$Ni_3GaSb$และ $Ni_3InSb$ให้เกิดปฏิกิริยาในหลอดซิลิกาที่ปิดสนิทที่อุณหภูมิสูง ผลการทดลองของสารประกอบ $Ni_3GaSb$และ $Ni_3InSb$ พบว่ามีค่า $\alpha$และ$\rho$ ที่มีสมบัติคล้ายโลหะโดยค่า power factor และ dimensionless figure of merit มีค่าเพิ่มขึ้นเมื่ออุณหภูมิสูงขึ้น และมีค่าสูงสุดที่ 1073 Kโดยdimensionless figure of merit ของ $Ni_3GaSb$ และ $Ni_3InSb$ มีค่าเท่ากับ 0.022 และ 0.023 ตามลำดับ

# TABLE OF CONTENTS

# LIST OF TABLES

# LIST OF FIGURES

# LIST OF APPENDIX TABLE

## LIST OF APPENDIX FIGURES

## ABBREVIATIONS AND SYMBOLS

| | | |
|---|---|---|
| RF | = | Radio frequency |
| kW | = | Kilowatts |
| AC | = | Alternating current |
| DC | = | Direct current |
| $^{o}C$ | = | Degree celcius |
| cm | = | Centimeter |
| mm | = | Millimeter |
| μm | = | Micrometer |
| nm | = | Nanometer |
| Å | = | Angstrom |
| EDX | = | Energy dispersive X-ray spectroscopy |
| FT-IR | = | Fourier-transform infrared spectrometry |
| PL | = | Photoluminescence spectrometry |
| SEM | = | Scanning electron microscopy |
| TEM | = | Transmission electron microscopy |
| XRD | = | X-ray diffraction spectrometer |
| JCPDS | = | The Joint Committee for Powder Diffraction Standards |
| SPS | = | Spark plasma sintering |
| $Z$ | = | Figure of merit |
| $ZT$ | = | Dimensionless figure of merit |
| TE | = | Thermoelectric |

| | | |
|---|---|---|
| $C_p$ | = | Heat capacity |
| $D$ | = | Crystallite size |
| $K$ | = | Geometric (shaped) factor |
| $E_{el}$ | = | The electric field strength |
| $E_g$ | = | Energy band gap |
| $N$ | = | Number of atoms in the unit cell |
| $R$ | = | Gas constant |
| $T$ | = | Temperature |
| $T_H$ | = | Hot-site temperature |
| $T_C$ | = | Cold-site temperature |
| $V$ | = | Unit cell volume |
| $b$ | = | The path length |
| $\alpha$ | = | Seebeck coefficients |
| $\rho$ | = | Electrical resistivity |
| $\sigma$ | = | Electrical conductivity |
| $\kappa$ | = | Thermal conductivity |
| $\kappa_{lat}$ | = | Lattice thermal conductivity |
| $\kappa_{el}$ | = | Electronic thermal conductivity |
| $\kappa_{dif}$ | = | Thermal diffusivity |
| $\alpha_{abs}$ | = | Total absorption coefficient |
| $k_B$ | = | Boltzmann constant |
| $k$ | = | The rate constant of reaction |
| $h$ | = | Planck's constant |

| | | |
|---|---|---|
| $t$ | = | Time |
| $x$ | = | The amount of reaction |
| $v$ | = | Photon frequency |
| $d$ | = | Interplanar planes |
| $d$ | = | Sample density |
| $d_{th}$ | = | Theoretical density |
| $d_{exp}$ | = | Measured density |
| $d_{exp/dexp}$ | = | Relative density |
| $n_s$ | = | Sheet carrier density |
| $n_H$ | = | Hall carrier concentration |
| $V_H$ | = | Hall voltage |
| $I$ | = | Current |
| $B$ | = | Constant magnetic field |
| $q$ | = | Elementary charge |
| $R_H$ | = | Hall coefficient |
| $\mu_H$ | = | Hall mobility |
| $\Phi$ | = | Coefficient of performance |
| $\eta$ | = | Efficiency |
| $W$ | = | Power input |
| $Q_H$ | = | Net heat flow rate |
| $\omega$ | = | Angular frequency |
| $\varepsilon_r''$ | = | The imaginary part of the complex relative permittivity |
| $\varepsilon_0$ | = | The permittivity of free space |