SILICON NANOWIRE ARRAYS FOR THERMOELECTRIC POWER HARVESTING

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I dedicate this thesis to my beloved parents,
siblings and friends who always there for me.
Thank you for your full support.
May Allah bless them all.

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ABSTRACT

Numerous types of thermoelectric materials with best thermoelectric performances have been explored such as bismuth-telluride (Bi_2Te_3), which is the most commonly found in the market, has a figure-of-merit close to one. However, due to limited sources, highly toxic and expensive, the application of one-dimensional nanomaterial is proposed in thermoelectric micro-energy harvesting, which has been predicted to show improvement in thermoelectric properties. Use of Silicon Nanowire Arrays (SiNWA) as thermoelectric material was reported to reduce thermal conductivity, κ, by a hundredfold compared to bulk Silicon (Si). The properties such as heat flow, temperature difference, ΔT between hot and cold junctions and Seebeck voltage, V_{oc} were evaluated concurrently for different lengths of p- and n-type SiNWA. This thesis reports the performance of SiNWA with two different lengths, 30 µm and 50 µm, on both p- and n-type Si for thermoelectric energy harvesting, and followed by comparing the recorded performance to its bulk Si. A simple and cost-effective technique, metal-assisted chemical etching (MACE), was used to fabricate SiNWA and the nanowires lengths were characterized. An increase in thermal resistance reduces κ for Si, which is advantageous for a thermoelectric material. In this work, heat flow was noticeably decreased in SiNWA samples, resulting in a higher ΔT and V_{oc} than in bulk Si. A larger ΔT between junctions is also attainable in SiNWA by increasing nanowires length. The results have shown that both p- and n-type SiNWA samples (50 µm) have achieved 95 % and 96 % increases in ΔT, respectively, relative to bulk Si samples. In addition, as the length of nanowires increased, a longer time was required to reach a steady value of ΔT. The reduction on approximation values of κ by a hundred-fold which increases thermal resistance as well as Seebeck coefficient, S in the SiNWA samples. Improvement in SiNWA thermoelectric properties will expands the application of SiNWA thermoelectric micro-energy harvesters in various fields such as bio-medical, telecommunication, wireless technologies and others.

ABSTRAK

Pelbagai jenis bahan termoelektrik dengan prestasi termoelektrik yang terbaik telah diterokai seperti bismut telurida (Bi_2Te_3) yang terdapat di pasaran dan memiliki angka-merit menghampiri nilai satu. Walau bagaimanapun, disebabkan bahan ini yang mempunyai sumber yang terhad, bertoksik tinggi dan mahal, bahan nano satu-dimensi dicadangkan untuk kegunaan dalam penuaian tenaga mikro termoelektrik yang mana telah diramalkan dapat menunjukkan peningkatan dalam sifat termoelektrik. Penggunaan jajaran nano-wayar silikon (SiNWA) sebagai bahan termoelektrik dilaporkan telah dapat mengurangkan daya pengaliran haba, κ, sebanyak seratus kali ganda berbanding Si pukal. Sifat-sifat seperti aliran haba, perbezaan suhu, ΔT diantara simpang panas dan sejuk serta voltan Seebeck, V_{oc} telah dinilai secara serentak bagi SiNWA jenis p- dan n- bagi kepanjangan nano-wayar yang berbeza. Tesis ini melaporkan prestasi SiNWA dengan dua panjang nano-wayar yang berbeza iaitu, 30 µm dan 50 µm bagi kedua-dua jenis p- dan n- Si untuk penuaian tenaga termoelektrik, dibandingkan dengan prestasi yang direkod oleh Si pukal. Satu teknik mudah dan kos efektif iaitu punaran kimia berbantu logam (MACE), telah digunakan untuk membentuk SiNWA dan panjang nano-wayar yang terhasil telah dikenalpasti. Peningkatan dalam rintangan haba dapat mengurangkan κ bagi Si, yang merupakan salah satu ciri terbaik bagi bahan termoelektrik. Dalam kerja ini, aliran haba ternyata berkurangan secara ketara bagi sampel SiNWA, yang berupaya untuk menghasilkan ΔT dan V_{oc} yang lebih tinggi berbanding Si pukal. ΔT yang lebih besar antara simpang panas dan sejuk juga dapat dicapai dalam sampel SiNWA dengan menambahkan panjang nano-wayar. Hasil kajian menunjukkan bahawa kedua-dua sampel SiNWA (50 um) jenis p- dan n-, masing-masing mencapai 95 % dan 96 % peningkatan dalam ΔT, berbanding sampel Si pukal. Selain itu, semakin bertambah panjang nano-wayar, semakin lama masa yang diperlukan untuk mencapai nilai mantap bagi ΔT. Anggaran nilai κ di dalam kajian in berjaya dikurangkan sebanyak seratus kali ganda, di mana dapat membantu untuk meningkatkan rintangan haba dan nilai pekali Seebeck, S dalam sample SiNWA berbanding Si pukal. Kemajuan dalam sifat termoelektrik bagi bahan SiNWA dapat memperluaskan penggunaannya di dalam pelbagai bidang seperti bio-perubatan, telekomunikasi, teknologi tanpa wayar dan sebagainya.

TABLE OF CONTENT

LIST OF FIGURES

LIST OF TABLES

LIST OF ABBREVIATIONS

°C - Degree Celsius
µm - Micrometre
µW - Microwatt
AFM - Atomic Force Microscopy
Ag - Silver
Ag^+ - Silver ion
$AgNO_3$ - Silver Nitrate
Al - Aluminium
Au - Gold
Ba - Barium
Bi - Bismuth
Bi_2Te_3 - Bismuth Telluride
BOE - Buffered Oxide Etchant
cm^2 - Square centimetre
cm^3 - Cubic centimetre
CMOS - Complementary Metal-Oxide-Semiconductor
Cu - Copper
CVD - Chemical vapor deposition
DC - Direct current
DI - Deionized
DRIE - Deep reactive ion etching
EE - Electroless etching
EM - Electromagnetic

FEA	-	Finite Element Analysis
FESEM	-	Field Emission Scanning Electron Microscope
H^+	-	Hydrogen ion
H_2O	-	water
H_2O_2	-	Hydrogen Peroxide
HF	-	Hydrofluoric Acid
i.e.	-	that is
ICP	-	Inductive-coupled plasma
IR	-	Infrared
K	-	Kelvin (Temperature)
K	-	Potassium
LAN	-	Local Area Network
MACE	-	Metal-Assisted Chemical Etching
MBE	-	Molecular beam epitaxy
MEMS	-	Microelectromechanical system
min	-	Minute
mm	-	Millimetre
Mo	-	Molybdenum
MP	-	Megapixel
mV	-	millivolt
mW	-	milliwatts
Na	-	Sodium
nm	-	Nanometre
Pb	-	Lead
Poly-Si	-	Polysilicon
RFID	-	Radio Frequency Identification
Sb	-	Antimony
Sb_2Te_3	-	Antimony Telluride
sec	-	Second

Si - Silicon

Si^{4+} - Silicon ion

SiNW - Silicon Nanowire

SiNWA - Silicon Nanowire Arrays

SiO_2 - Silicon Dioxide

SnSe - Tin Selenide

TEG - Thermoelectric Generator

VLS - Vapor-Liquid-Solid

LIST OF SYMBOLS

ΔT	-	Temperature difference
C_p	-	Heat capacity at constant pressure
D	-	Density
E°	-	Electrochemical potential
j	-	Current density
l_Cu1	-	Length of top Cu sheet
l_Cu2	-	Length of bottom Cu sheet
l_Si	-	Length of Si substrate underneath the nanowires
L_Si	-	Length of Si nanowire
Q	-	Heat flow, Watts
$R_\mathrm{c,l}$	-	Thermal contact resistances of the lower interfaces between Si sample and bottom Cu sheet and
$R_\mathrm{c,total}$	-	Total thermal contact resistance
$R_\mathrm{c,u}$	-	Thermal contact resistances of the upper interfaces between top Cu sheet and Si sample
S	-	Seebeck coefficient
S_nw	-	Seebeck coefficient of nanowire
T	-	Absolute temperature
T_cold	-	Temperature at cold junction
T_Cu1	-	Temperature of top Cu sheet
T_hot	-	Temperature at hot junction

T_S - Temperature of the Si sample

V - Potential difference

V_oc - Open-circuit voltage/Seebeck voltage

ZT - Dimensionless Figure-of-Merit

ε_r - Relative permittivity

κ - Thermal conductivity

κ_air - Thermal conductivity of air

κ_Cu - Thermal conductivity of Cu

κ_e - Electronic thermal conductivity

κ_l - Lattice thermal conductivity

κ_nw - Thermal conductivity of nanowire

κ_Si - Thermal conductivity of Si

κ_Total - Total thermal conductivity

ρ - Resistivity

ρ_nw - Resistivity of nanowire

σ - Electrical conductivity

$\boldsymbol{\nabla} T$ - Temperature gradient

$\boldsymbol{\nabla} V$ - Voltage gradient

LIST OF APPENDICES

CHAPTER 1

INTRODUCTION

1.1 Research Background

Battery or button cells are the most common electrical energy source for low-power and portable electronic applications such as smartphones, embedded devices, remote sensors and medical implants. However, the limited lifespan of these energy sources affects device performance and the battery must be replaced periodically. As an alternative, energy harvesting technologies that capture and convert energy available in the surroundings into usable electrical energy could give limitless operating life for low-power devices, eliminating batteries replacement that is expensive, impractical and risky. There are various types of useful energy available in the environment such as light, kinetic, thermal and electromagnetic energy, which can be converted into electrical energy. Energy harvesters such as photovoltaic cells, piezoelectric transducers and thermoelectric generators are able to convert these useful energies to power up any low-power portable electronic devices. Most energy harvesters are designed to be cost-effective and require minimal maintenance over lifespans of several years. Some of energy harvesters are built in a miniature size which allowing them to be embedded in electronic devices.

A thermoelectric device is an inexpensive energy harvester that can be used to convert heat from the ambient, human body and waste heat into useful electrical energy. This type of energy harvester has a solid-state mechanism that actually increases the longevity of the device, while ensuring emissions- and noise-free operation, consequently promoting a healthy environment [1]. This type of energy harvester is currently used in several applications including wristwatches [2], biometric sensors [3], and cooling chambers [4].

1.2 Problem Statement

Various types of thermoelectric materials with excellent thermoelectric performances have recently been explored and reported [5-8]. Most thermoelectric devices found in the current market are made of bismuth-telluride (Bi_2Te_3), which exhibits a figure-of-merit (*ZT*) of close to one [9]. Although this compound is commonly used for thermoelectric devices, this material contain high toxic level and expensive due to its rarity. Alternatively, Si, a semiconductor material that is extensively used in microelectronic devices became a promising material for application in thermoelectric devices. However, the high thermal conductivity, κ, of bulk Si (i.e., ~150 $Wm^{-1}K^{-1}$ at room temperature [10, 11]) contributes to a low *ZT*, which consequently affects thermoelectric efficiency.

To address the shortcomings of existing materials, nanostructured materials such as nanowires [12, 13] and nanotubes [14-16] were proposed as an alternative to enhance thermoelectric properties [17]. Nanowires-based materials offer various advantages for improving thermoelectric performances, including thin-film superlattices and quantum dot-based materials [18]. Numerous studies in the literature have reported a hundredfold reduction in κ by using the Si nanowires [19-21]. Reduction of κ is due to frequent phonon scattering in the material which consequently improves the values of *ZT* which relates to the efficiency of thermoelectric devices [22, 23].

Despite the existing literatures on thermoelectric materials, knowledge gaps remain. In this work, the length of Si nanowire arrays (SiNWA), heat flow and temperature difference, ΔT between the hot and cold junctions, which would contribute improvements in output power, current and voltage of a SiNWA thermoelectric energy harvester, are studied together for both p-type (boron-doped) and n-type (phosphorus-doped) samples. The SiNWA are fabricated using a simple and cost-effective technique called metal-assisted chemical etching (MACE). Improvements that would portrayed by SiNWA as a potential thermoelectric material could open up more possibilities in thermoelectric applications.

1.3 Research Objectives

Thermoelectric energy harvester that uses temperature gradients and heat flow present in nature and human body offers a method to overcome the problems. However, thermoelectric material plays an important role in producing efficient thermoelectric devices. In this work, a nanostructured SiNWA is proposed as an alternative thermoelectric material. Therefore, the main objectives of this research include:

1) To develop several lengths of p- and n-type Si Nanowire Arrays (SiNWA) thermoelectric material in the range of 5 – 50 μm by using MACE technique.

2) To measure and characterize thermoelectric properties such as heat flow, ΔT and output voltage of p- and n-type SiNWA thermoelectric samples.

1.4 Scope of Research

The scopes of this work are as follow:

i. This research aims to explore a one-dimensional nanostructured semiconductor material, SiNWA which is used as a thermoelectric material. Two types of Si wafers were used in this work, namely p-type (boron-doped) and n-type (phosphorus-doped).

ii. Simulations of heat transfer and output voltage between hot and cold ends of a single Si nanowire model were conducted while bulk Si model was used as a benchmark. Simulations were done in COMSOL Multiphysics software.

iii. Wet etching technique, metal-assisted chemical etching (MACE) was used to fabricate nanowires. Several lengths of p- and n-type Si Nanowire Arrays (SiNWA) thermoelectric material in the range of 5 – 50 µm were fabricated in order to analyse their relationship to the time of etching.

iv. The amount of heat applied to the hot junction by the heater was assumed the same in all the experiments, where the heating rate was 0.5 °C/sec. Experiments in this work were conducted to evaluate the performance of the SiNWA samples during temperature changes.

1.5 Research Contribution

Nanostructured SiNWA was predicted to show an improvement in thermal resistance due to enhancement of thermoelectric properties such as ΔT and Seebeck voltage, V_{oc} compared to bulk Si. Both p- and n-type SiNWA were developed using MACE wet etching technique where the longer etching time will increases the length of SiNWA. Heat flow, ΔT and V_{oc} are characterized simultaneously and analysed for two different nanowire lengths, using bulk Si for comparison. Heat flow across SiNWA was expected to be reduced which will be able to provide a larger ΔT between the hot and cold junctions than bulk Si. A larger ΔT that can be attained in a thermoelectric device may help to gain a larger V_{oc} and finally, improves thermoelectric performance. Improvement of V_{oc}, heat flow and ΔT across SiNWA will expand the use of SiNWA in thermoelectric devices for applications such as industrial monitoring, electrical appliances and others.

1.6 Potential Impact of the Research

Thermoelectric energy harvesting offers a promising self-sustainable source of energy for low-power applications. Si is extensively used in microelectronic devices and is a promising material in thermoelectric devices. Previous studies have shown that by altering bulk Si into nanostructured Si such as SiNWA improves thermoelectric properties. The use of SiNWA could therefore be profitable in thermoelectric applications. The output voltage from a SiNWA can be varied or increased by adjusting nanowires length, thereby changing ΔT. This could be beneficial for certain low-power applications.

Results of this study could motivate other researchers to further explore the application of SiNWA to thermoelectric energy harvesters. By reducing manufacturing and material costs, the use of thermoelectric devices could potentially be expanded into various fields such as automotive, bio-medical engineering and wireless technologies. In addition, cost reductions may improve economic growth and perhaps encourage the use of green technology in our daily lives for environmental conservation.

1.7 Organization of the Thesis

This study focuses on the use of SiNWA as a thermoelectric material and how different lengths of nanowires affect the output of a thermoelectric device. All processes in the study are described in this thesis. This thesis consists of five chapters starting with an introduction of research in Chapter 1. This chapter includes background, problem statements, objectives, scopes, contribution and potential impacts of the research.

Chapter 2 provides an overview of different types of energy harvesters. The fundamental working principles of thermoelectric energy harvester are described. Additionally, previous work on thermoelectric materials is reviewed. Finally, the theory and significance of SiNWA as a thermoelectric material are reviewed at the end chapter.

Chapter 3 explains the research methodology, including workflow and models used for simulations of heat distribution over nanowires. Fabrication of SiNWA using a two-step MACE technique is described in this chapter. In addition, experimental setup and equipment used in the experiment are described at the end of the chapter.

Chapter 4 presents the experimental analysis of SiNWA as a thermoelectric material. This chapter analyses and discusses three results sections: COMSOL simulations, MACE fabrication and experimental characterization.

Finally, the findings of this work are summarized in Chapter 5. Recommendations for future work are given to help others further develop this technology and improves the output of this work.

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