CHAPTER 1

INTRODUCTION

1.1 Background of Study

Thermoelectricity is a term that refers to a process where heat energy is converted to electrical energy or vice versa. The processes are conducted by using thermoelectric materials due to the three main effects involved in the process namely as Seebeck effect, Peltier effect, and Thomson effect. The device made of this thermoelectric material is widely known as a promising technology for electric power generation and refrigeration in various fields. Nowadays, the most promising thermoelectric application is generating power from waste heat (S N Mustafa et al., 2017).

Performance is determined by its dimensionless figure-of-merit, *ZT* which can be calculated using the formula,

$$ZT = \frac{\alpha^2}{\rho\lambda}T \tag{1.1}$$

where α, T, λ, and ρ are Seebeck coefficient, absolute temperature, thermal conductivity, and electrical resistivity respectively. A good thermoelectric material should have a high Seebeck coefficient, α but low electrical resistivity, ρ, and thermal conductivity, λ. Higher *ZT* indicates the higher performance of thermoelectric materials as a converter. However, higher *ZT* is not easily obtained. Thus, a lot of studies have been conducted to improve *ZT*. Among those, thermoelectric materials and their structure play an important role in improving TE performance. State-of-the-art TE

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materials are Bi_2Te_3, PbTe, and SiGe which operate at the different optimum temperatures of 300-400 K, 500-900 K, and above 900 K respectively. These bulk TE properties can be enhanced by tuning or doping techniques (Alam et al., 2013).

On the other hand, thermoelectric structures have been widely studied and recent progress in ZT shows great assurance in the development of a new generation thermoelectric generator (*TEG*) in cascade and segmented structures (Shen, Liu, Wu S., Xiao, & Chen, 2018)(Ouyang, 2016). In this new structure, different materials were segmented together, for example, Bi_2Te_3 and PbTe so that the thermoelectric leg can operate at a wider temperature range of 300 K – 900 K.

Another way to improve thermoelectric properties is through fabrication techniques. There were several techniques in fabricating thermoelectric materials which involved physical and chemical methods. Sintering methods are frequently used to fabricate TE materials. Sintering methods include different techniques such as pressureless sintering, hot isostatic pressing (HIP), hot pressing (HP), and spark plasma sintering (SPS). The most reliable sintering method is using the spark plasma sintering (SPS) technique also known as the field-assisted sintering technique (FAST). The difference between this technique from other sintering techniques is that the SPS technique could fabricate the materials in a short period with the presence of electric current. This method fastens the sintering process where the heating rate is up to 1000 degrees per minute. According to Siebert et al. (2019), the rapidly increasing temperature of the SPS technique allows the densification to take place in a low-temperature state under a short time process. To be exact, fast densification can be achieved as grain growth can be avoided. It is said that the increase in heat rate decreased the grain growth. Meanwhile, the presence of electrical current generates

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plasma which produces an extreme increase in temperature on the surface (T. Hungria et al., 2009).

1.2 Problem Statement

Not much study has been made on the fabrication of bulk thermoelectric using another sintering method due to a well-established study. Researchers are more focused on the fabrication of thin-film and green material which has been trending nowadays. However, it could not be denied that state-of-the-art bulk thermoelectric devices still outperform thermoelectric thin film and has wider application.

Common fabrication of thermoelectric materials using the SPS technique is not available in Malaysia where the previous researcher has sent the samples to be fabricated outside Malaysia (Rizwan M. et. al, 2020). Thus, the study on bulk thermoelectric material is limited due to the high cost of thermoelectric fabrication. An alternative approach for low-cost construction of bulk thermoelectric materials is using the HP fabrication technique. This is due to the cheaper hot press machine and widely available in Malaysia. Therefore, this study, resorts to using HP techniques, to study the optimum thermoelectric fabrication technique. Since the HP technique tends to produce less efficient thermoelectric materials, this study used state of art thermoelectric material, Bi_2Te_3- and PbTe-based.

In addition, there is a limitation in knowledge about the procedure for preparing the mass of materials needed in previous studies. Although the mass of powder can be measured according to the stoichiometry ratio. The outcome might be affected by the fabrication process. Ability to identify the correct measurement of the materials would ease problems such as waste of material. It is indeed crucial to initially study the methods to prepare samples. Hence, this study will investigate 2 different techniques to prepare the sample using the HP process.

Another problem involves TE performance. Since the real condition that thermoelectric experienced is under large temperature differences, the properties of thermoelectric such as the Seebeck coefficient need to analyze under such conditions. However, most previous research investigated the properties under small temperature differences (Abrutin,2004) (Zhao et.al, 2014) (Ouyang, 2016). Analyzing the properties under a small temperature difference doesn't necessarily provide a real condition for the measurement of the properties which is under a large temperature difference. Therefore, this study attempts to measure and compare the Seebeck coefficient under these two conditions, large and small temperature differences. As there is no commercial equipment in measuring the properties under large temperature differences, a high-temperature vacuum chamber is created to overcome the issue.

1.3 Research Objectives

The objectives of this study are as follows:

1. To fabricate an alternative cost-effective thermoelectric material (PbTe- and Bi_2Te_3-based) using a hot press method based on two preparation techniques which are atomic ratio and volume ratio method.
2. To design and fabricate the in-house vacuum chamber system for high-temperature measurements up to 1000 K.

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3. To measure the Seebeck coefficient under large temperature difference (lΔT) using in-house apparatus inside and outside vacuum chamber and electrical resistivity using a 4-point probe.
4. To compare the Seebeck coefficient under large (lΔT) and small temperature difference (sΔT) to see the difference between both conditions.

1.4 Scope/Limitation of the Study

In this study, the thermoelectric material samples used are limited to n- and p-type BiTe-based and PbTe-based material with a composition of $Bi_2Te_{2.7}Se_{0.3}$, $Bi_{0.3}Sb_{1.7}Te_3$, $PbTe_{0.8}Se_{0.2}$, and $Pb_{0.4}Sn_{0.6}Te$. Thermoelements were constructed using the hot press method with a limiting temperature of the hot press machine of 573 K. Limited pressure of the automatic hot press machine is 13 MPa.

Since the Seebeck coefficient under large temperature difference (lΔT) will be measured under this research, and no available commercial equipment available, a refurbished homemade device was developed. Limitation for this equipment, allowing only measurement under large temperature differences. Although the limitation of the heater is increased up to 1173 K from the previous design, the hot side can only be heated up to about 500 K.

Electrical resistivity measurement will be conducted at room temperature only using a 4-point probe. Besides, the 4-point probe measured the resistivity on the surface of samples.