Preparation of n-type Bi2Te3 Films by Electrophoretic Deposition

TALEBI, Tahereh, GHOMASHCHI, Reza, TALEMI, Pejman and AMINORROAYA YAMINI, Sima <http://orcid.org/0000-0002-2312-8272>

Preparation of n-type Bi_2Te_3 Films by Electrophoretic Deposition

Tahereh Talebi, Reza Ghomashchi, Pejman Talemi, Sima Aminorroaya

***Abstract*—**A high quality crack-free film of Bi_2Te_3 material has been deposited for the first time using electrophoretic deposition (EPD) and microstructures of various films have been investigated. One of the most important thermoelectric (TE) applications is Bi_2Te_3 to manufacture TE generators (TEG) which can convert waste heat into electricity targeting the global warming issue. However, the high cost of the manufacturing process of TEGs keeps them expensive and out of reach for commercialization. Therefore, utilizing EPD as a simple and cost-effective method will open new opportunities for TEG's commercialization. This method has been recently used for advanced materials such as microelectronics and has attracted a lot of attention from both scientists and industry. In this study, the effect of media of suspensions has been investigated on the quality of the deposited films as well as their microstructure. In summary, finding an appropriate suspension is a critical step for a successful EPD process and has an important effect on both the film's quality and its future properties.

***Keywords*—**Bi_2Te_3, electrical conductivity, electrophoretic deposition, thermoelectric materials, thick films.

I. Introduction

GLOBAL energy consumption is increasing rapidly. To ensure a sustainable future, there is a need to develop better ways to produce and utilise energy. Almost all of the energy production methods, as well as industrial processes, generate heat that cannot be used and will be wasted and released into the atmosphere, increasing the threat of global warming. Nearly 60% of the energy produced by mankind is in the form of wasted thermal energy [1]. If this enormous amount of energy could be harnessed into more useful energy forms, it would reduce greenhouse gases and fossil fuel consumption, significantly reducing the risks of global warming [2], [3].

The solution for this problem may lie in using TE materials, which are capable of converting waste heat into electricity. The waste heat in systems such as power plants, cars, planes, computers or even stovetops can be sources of electricity. TE materials could also be used for refrigeration purposes, without the shortcomings associated with vibration in conventional chillers [4], [5].

When a thermal gradient is applied to TE materials, it will generate electricity. The efficiency of this conversion relies on the magnitude of the temperature difference, and intrinsic properties of the TE materials, as well as the design of the TE generator in terms of its shape, electric contact, etc. [2].

Before the commercialization of these devices, various challenges have to be solved by researches and industry to increase their uses from limited niche applications, such as deep-space satellites, high temperature waste heat recovery and remote power generation for unmanned systems [1], [6]. Two of the most important challenges are the high weight of conventional TE devices and the high cost of manufacturing them [1], [7], [8].

To lower the weight of these devices, especially for mobile applications, fabricating TE films has been the focus of current research in the industry [9]. TE films can have interesting and varied applications. For instance, body heat can be harnessed to recharge a mobile phone, iPad or other personal gadgets [10]. Therefore, the feasibility of TE devices heavily depends on the cost and energy conversion efficiency. Taking new approaches toward TE materials using nanostructures and novel crystal structures has resulted in a rapid development of TE materials with significant higher energy efficiencies [1], [5], [12].

Thermoelectric materials have various classifications such as Half-Heusler materials, Clathrates, skutterudites, Chalcogenides which have different characteristics [2], [7], [16].

Bismuth Telluride, the most studied TE material, is a simple TE material which is commercially available and has interesting TE properties.

Fig. 1 shows the Bi-Te binary phase diagram [4]. The arrow shows the position of Bi_2Te_3 in the diagram. Its Seebeck coefficient depends on its composition (Te deficiency) and doping materials. The maximum ZT values reported for pure crystalline Bi_2Te_3 materials are about 0.75 and 0.86 for p-type and n-type, respectively [11].

As an advantage, a complete solid solubility exists among Bi_2Te_3, Sb_2Te_3 and Bi_2Se_3 which all have noble TE features. Therefore, ZT and other TE properties can be improved by the addition of Sb_2Te_3 and Bi_2Se_3 to Bi_2Te_3 [3]. Bi_2Te_3 can form p-type and n-type semiconductor solid solutions simply by adding Sb_2Te_3 or Bi_2Se_3, resulting in $Bi_{2-x}Sb_xTe_3$ and $Bi_2Te_{3-x}Se_x$ alloys, respectively [5]. This will result in lowering thermal conductivity but not degrading electrical conductivity. The optimum compositions with ZT~1 at room temperature

T. Talebi is with the Mechanical Engineering School, the University of Adelaide, South Australia, Australia (phone: 883-133-360; fax: 883-134-367; e-mail: tahereh.talebinamakrodbari@adelaide.edu.au).

R. Ghomashchi is with the Mechanical Engineering School, the University of Adelaide, South Australia, Australia (e-mail: reza.ghomashchi@adelaide.edu.au).

P. Talemi is with the Chemical Engineering School, the University of Adelaide, South Australia, Australia (e-mail: pejman.talemi@adelaide.edu.au).

S. Aminorroaya is with Institute for Superconducting & Electronic Materials, University of Wollongong, New South Wales, Australia (e-mail: sima@uow.edu.au).

are usually $Bi_2Te_{2.7}Se_{0.3}$ (n-type) and $Bi_{0.5}Sb_{1.5}Te_3$ (p-type) [4], [8].

To decrease the cost of production, a cost-effective film fabrication method, called EPD has been used. This method is a simple non-vacuum technique with a high rate of deposition method to prepare thick films [11], [12].

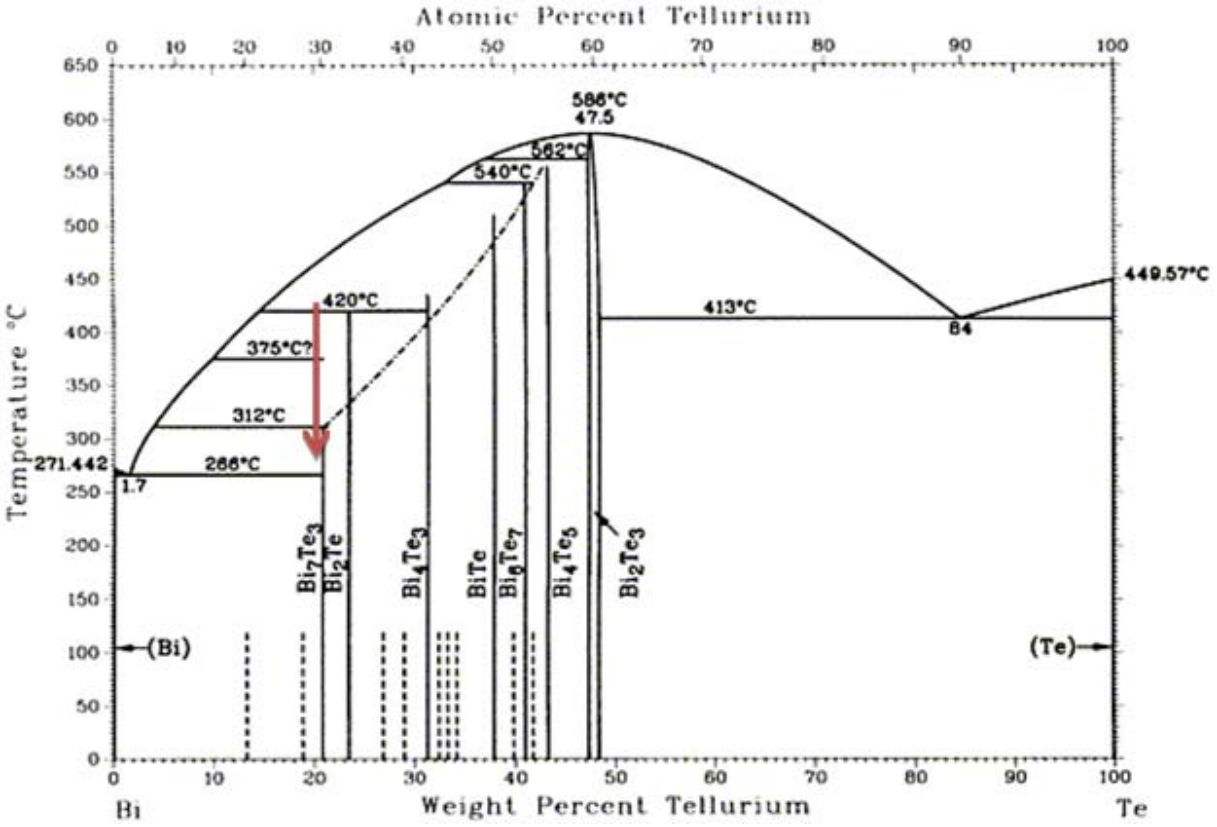


Fig. 1 The Bi-Te binary phase diagram [4]

In EPD, a DC current electric field is applied to a suspension of the charged powder particles, and then they are attracted and deposited on the substrate with the opposite charge. Different factors from both the suspension and EPD process can influence the deposited film [11], [13]. EPD has been used with numerous materials for a wide range of applications such as solid oxide fuel cells [14], functionally graded ceramics [15], superconductors [16] etc.

Determining the appropriate media for a suspension is a critical step to achieve a homogenous and dense film in EPD [17]. Therefore, in this research, various suspensions with different media have been prepared and the quality, the macrostructure and the microstructure of the EPD films have been investigated.

II. Experimental Procedure

Bi_2Te_3 powder was commercially available from the Bio-chemical Leader Company with 99.99% purity. The chemical composition of p-type Bi_2Te_3 powder was 22.51 Bi (wt %), 54.90 Te (wt %) and 23.59 Sb (wt %).

The median particle size (D50) of 20μm was characterized for particle sizes of the powder using a Malvern Mastersizer 2000 laser particle size analyser. Scanning electron microscope (SEM - Quanta 450) has been utilized to obtain the SEM images of the starting powder is shown in Fig. 2 at diverse magnifications.

To prepare the suspension, 1g of Bi_2Te_3 powders were dispersed in 100 ml of the media mixture (10 g/L Bi_2Te_3 suspension). The mixture was put in an ultrasonic bath for 15 min to disperse and break the agglomerates and stabilise the suspension. The suspension was then rested for at least for 2 hours to reach stability. The suspension was ultrasonicated for 2 minutes prior to the EPD process.

All of the media supplied by the Sigma Aldrich Company and the purity of the ethanol and ethylene glycol was 99.8%; the purity of the acetone and isopropanol was 99.5%; and the purity of the tetrahydrofuran and acetylacetone was 99.9%, 99%, respectively.

A schematic image of the EPD cell is shown in Fig. 3. The distance between the substrate and the counter electrode was constant at 10 mm for the EPD process. The electrodes were vertical and kept parallel and fixed during the EPD process.

The substrate and the counter electrode were cut from a commercially obtained copper foil roll, with dimensions of 10 mm by 10 mm.

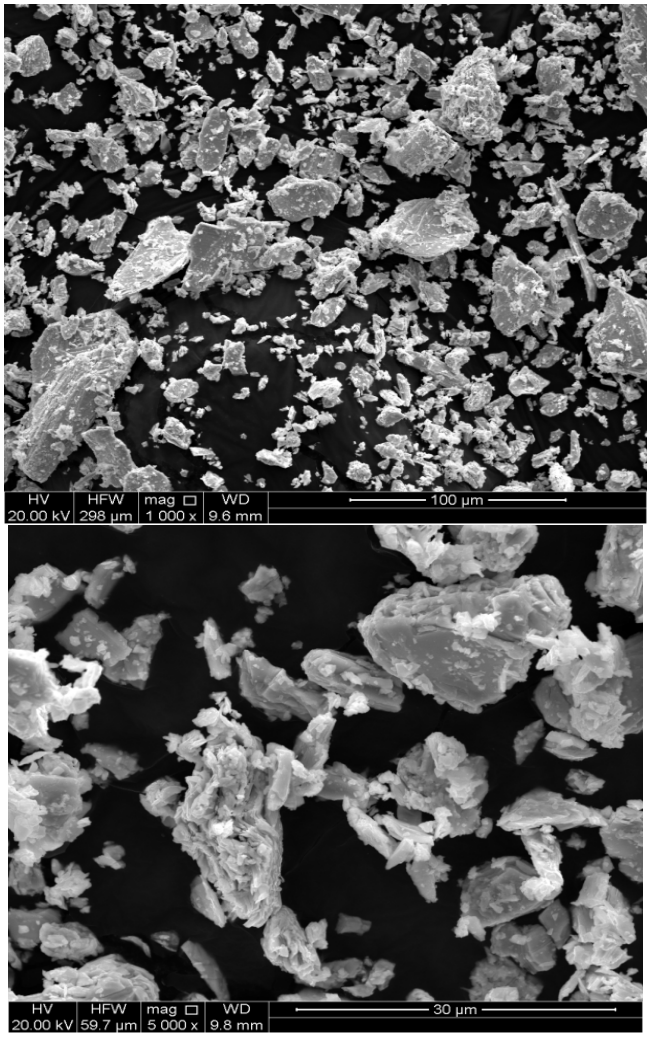


Fig. 2 The SEM image of p-type Bi_2Te_3 powder at diverse magnifications (1000x and 5000x)

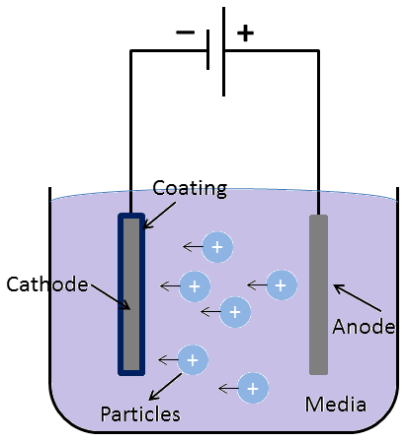


Fig. 3 Schematic image of the EPD cell

III. Results and Discussion

A film of Bi_2Te_3 has been successfully deposited on a copper substrate through modification of the suspension mixture. To achieve a high quality, uniform film, more adjustments and experiments were needed to reach a fundamental understanding of the effect of different parameters on the deposited films and their characteristics, as will be discussed in the following sections.

To reach the highest efficiency and figure of merit (ZT) for a TE material, the total thermal conductivity should be as low as possible. However, the thermal conductivity of metals is proportional to its electrical conductivity because it is dominated by electrons. The electrical conductivity of TE materials directly depends on its electronic thermal conductivity (k_e) which is a function of its band structure [1]. Thus, the lattice thermal conductivity (k_L) is the only parameter that can be reduced without too much negative effect on the electrical conductivity and figure of merit of TE materials. It is believed that phonons are responsible for transferring lattice thermal conductivity [3].

Thermal conductivity (k) consists of two of electronic (k_e) and lattice (k_L) components and T is the absolute temperature [6]. The power factor also has a significant effect on the performance of TE materials. The figure of merit can be increased by enhancing power factor and decreasing thermal conductivity. Semiconductors have the highest power factor amongst the materials because electrical conductivity of insulators and Seebeck coefficient of metals are too low [2].

Therefore, the mobility of phonons must be minimised whereas the mobility of electrons have to remain as high as possible to achieve the highest ZT value .It also has been suggested that the highest figure of merit will be accomplished in a material with the electrical properties of a crystal while it has the thermal properties of a glass which is known as the Phonon-Glass Electron-Crystal (PGEC) material [5].

A. The Effect of the Suspension on the Weight of Deposition

Fig 4 shows the deposition weights per area of suspensions with medium and high quality depositions at 100V for 9 minutes. The results show that the suspension with a higher zeta potential and electrophoretic mobility yielded a higher deposition weight, which is in good agreement with EPD theoretical principles.

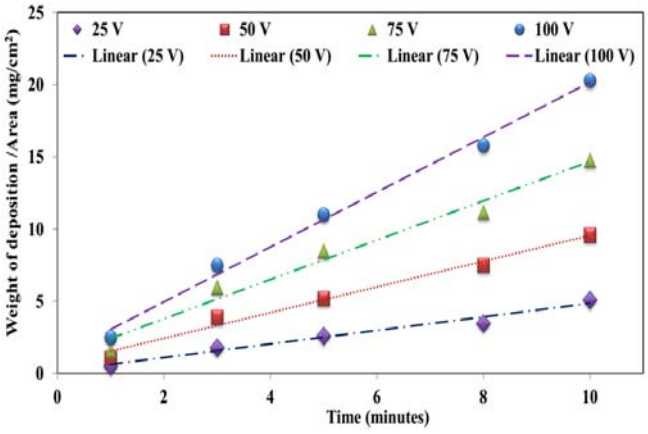
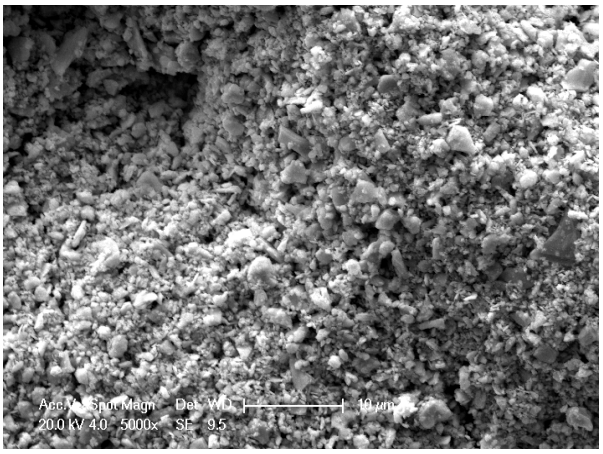


Fig. 4 The deposition weights per area of different suspensions with medium or high quality depositions.

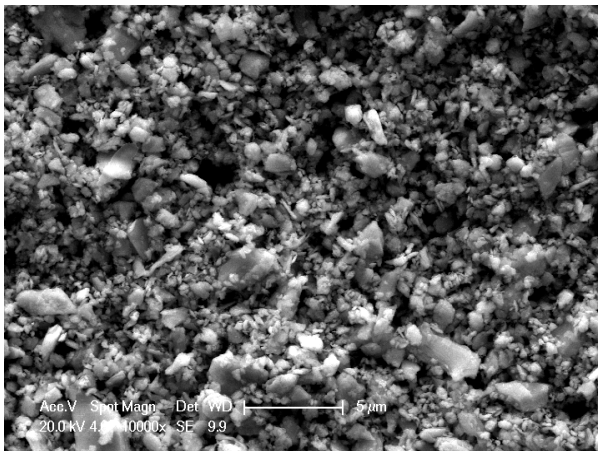
As discussed before, according to Hamaker's equation [11]-[13], the zeta potential and electrophoretic mobility have a direct dependence on the deposition weights. These relationships have also been confirmed by the experimental results, which show a similar trend for the deposition weights to the ones of zeta potential and electrophoretic mobility.

B. The Effect of EPD Time on the Film Microstructure

Fig. 5 I shows the SEM images of two green Bi_2Te_3 films deposited from a THF suspension at 100V/cm for (a) 5 minutes and (b) 10 minutes. It illustrates that when the deposition time doubles; the microstructure of the film also almost differs, which is in line with the measured results from the weight of the deposition experiments and Hamaker's equation as well as the EPD theoretical principle.



(a)



(b)

Fig. 5 The SEM images of green Bi_2Te_3 films at two different EPD voltages, (a) 100V and (b) 50V for 10 minutes from the 75% vol. Ethanol + 25% vol. Acetone suspensions

IV. Conclusion

In summary, a film of Bi_2Te_3 has been successfully deposited on a copper substrate and the effects of EPD time on the film microstructure as well as the effect of the suspension on the weight of deposition have been investigated. The result shows that time of the process and type of the suspension have significant effect on the deposited films.

Acknowledgment

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