Electrophoretic Deposition of p-Type Bi$_2$Te$_3$ for Thermoelectric Applications

Tahereh Talebi, Reza Ghomashchi, Pejman Talemi, Sima Aminorroaya

Abstract—Electrophoretic deposition (EPD) of p-type Bi$_2$Te$_3$ material has been accomplished, and a high quality crack-free thick film has been achieved for thermoelectric (TE) applications. TE generators (TEG) can convert waste heat into electricity, which can potentially solve global warming problems. However, TEG is expensive due to the high cost of materials, as well as the complex and expensive manufacturing process. EPD is a simple and cost-effective method which has been used recently for advanced applications. In EPD, when a DC electric field is applied to the charged powder particles suspended in a suspension, they are attracted and deposited on the substrate with the opposite charge. In this study, it has been shown that it is possible to prepare a TE film using the EPD method and potentially achieve high TE properties at low cost. The relationship between the deposition weight and the EPD-related process parameters, such as applied voltage and time, has been investigated and a linear dependence has been observed, which is in good agreement with the theoretical principles of EPD. A stable EPD suspension of p-type Bi$_2$Te$_3$ was prepared in a mixture of acetone-ethanol with triethanolamine as a stabilizer. To achieve a high quality homogenous film on a copper substrate, the optimum voltage and time of the EPD process was investigated. The morphology and microstructures of the green deposited films have been investigated using a scanning electron microscope (SEM). The green Bi$_2$Te$_3$ films have shown good adhesion to the substrate. In summary, this study has shown that not only EPD of p-type Bi$_2$Te$_3$ material is possible, but its thick film is of high quality for TE applications.

Keywords—Electrical conductivity, electrophoretic deposition, p-type Bi$_2$Te$_3$, thermoelectric materials, thick films.

I. INTRODUCTION

ALMOST 60% of the energy produced by mankind is wasted in the form of unusable thermal energy in various systems such as power plants, cars, planes, computers, or even stovetops. If we can utilise this enormous amount of energy, we can reduce fossil fuel consumption and greenhouse gases, as well help to decrease the effects of global warming [1], [2]. TE materials are especially attractive in this regard because they can convert waste heat into electricity. Using TE materials can increase the overall efficiency of the system by

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several percentage points [3], [4]. In addition, TEG are reliable and silent with no moving parts [5]. Although TEGs are currently being used in limited applications such as deep-space satellites, high temperature heat-waste recovery and remote power generation for unmanned systems, there remain a number of challenges which need to be addressed before the commercialization of these devices can occur [6], [7].

One of the challenges is the high production cost of TEGs. In addition, the devices are too heavy especially for mobile applications and their specific power to weight factor is low. Therefore, EPD as a cost-effective film fabrication method has been utilized to address these two vital issues for TE applications.

EPD is a film fabrication technique from colloidal suspensions [8]. In EPD, a DC electric field is applied to a suspension of charged particles enabling them to be attracted to the conductive substrate with the opposite charge and form a thick film [9].

EPD is a fast and cost-efficient method, requiring simple equipment. It has the ability to fabricate high quality and crack-free films with high degree of uniformity in the microstructure [10]. It is scalable for industrial applications and its setup can easily be adapted for complex-shaped substrates as well as flat, cylindrical, etc. [11]. This method has been used with numerous materials, for a wide range of applications such as hydroxyapatite for biomedical applications [12], electrolytes for solid oxide fuel cells [13], luminescent materials [14], functionally graded ceramics [15], carbon nanotubes [16], superconductors [17], nanosize zeolite membranes [18], etc. EPD also has industrial and medical applications such as corrosion protection processes.

In this paper, p-type Bi$_2$Te$_3$ films has been deposited using the EPD method, and a high quality crack-free thick film has been achieved for TE applications.

II. EXPERIMENTAL PROCEDURE

Bi$_2$Te$_3$ powder was commercially available from the Bio-chemical Leader Company with 99.99% purity. The chemical composition of p-type Bi$_2$Te$_3$ powder was 22.51 Bi (wt.%), 54.90 Te (wt.%) and 23.59 Sb (wt.%). The median particle size (D50) of 20 μm was characterized for particle sizes of the powder using a Malvern Mastersizer 2000 laser particle size analyser. The SEM image of the starting powder is shown in Fig. 1 at diverse magnifications.

A stable EPD suspension of p-type Bi$_2$Te$_3$ was prepared in a mixture of 25% Vol acetone + 75% Vol ethanol with 0.02g triethanolamine as a stabilizer for 100 ml of the media mixture. The purity of the acetone, ethanol, and

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triethanolamine supplied by the Sigma Aldrich Company was 99.5%, 99.8% and 99.5%, respectively.
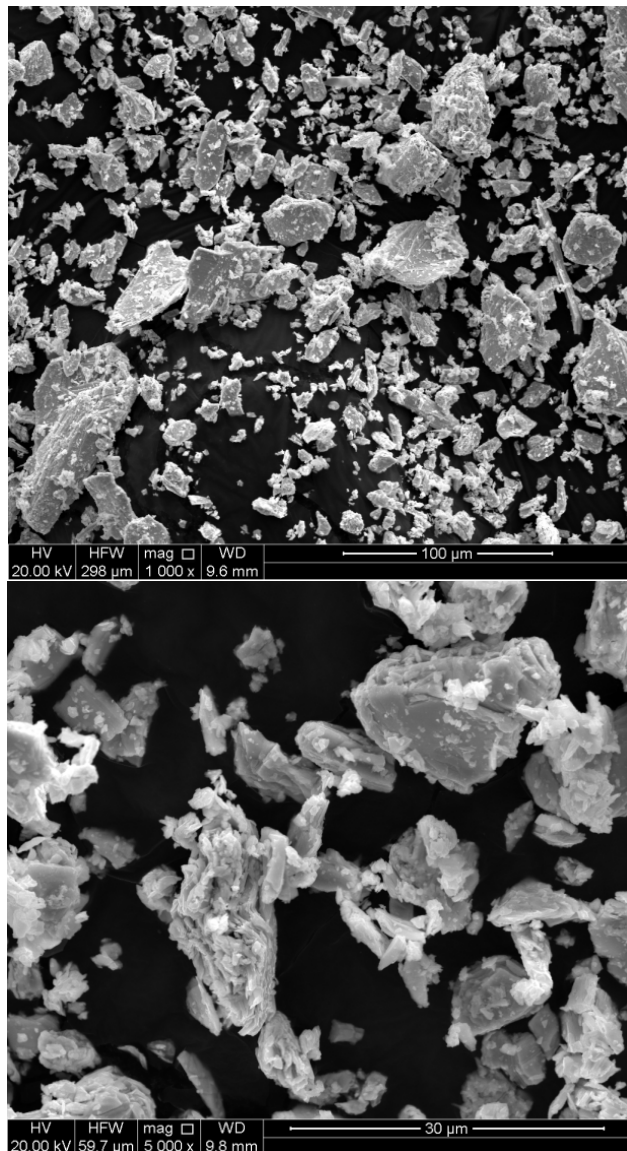
Fig. 1 The SEM image of p-type Bi_2Te_3 powder at diverse magnifications (1000x and 5000x)

To prepare the suspension, 1 g of Bi_2Te_3 powders was 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.

A schematic image of the EPD cell is shown in Fig. 2. 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*10 mm. A range of deposition voltages (0-100 V) and times (1-10 minutes) have been investigated for the EPD process.
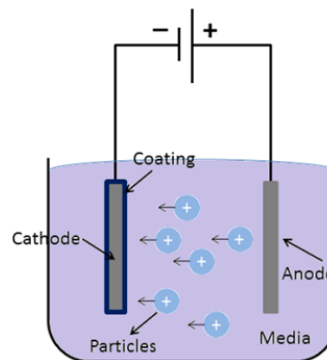

Fig. 2 Schematic image of the EPD cell

III. RESULTS AND DISCUSSION

A. The Effect of EPD Time and Voltage on the Deposition Weight

According to Hamaker's equation [19], the EPD deposit yield (w) is calculated through the following equation:

$$w = \int_{t_1}^{t_2} \mu . E . A . C . dt$$

where μ is the electrophoretic mobility; E is the electric field strength; A is the surface area of the electrode; C is the particle mass concentration and t is the deposition time.

Fig. 3 shows the deposition weights of the suspension at different applied voltages and deposition times. It shows that the deposition weights increase when the applied voltages or deposition times increase. It also shows that there is a linear dependence between these two EPD factors and the deposition weights, which is in good agreement with Hamaker's equation and EPD theoretical principles.
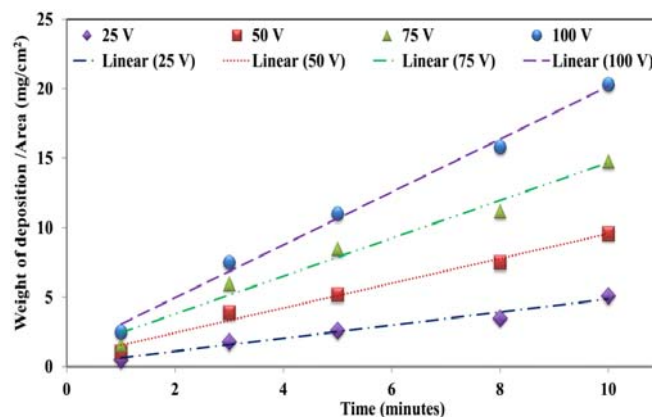

Fig. 3 The linear dependence between the EPD time and voltage with the deposition weight

B. The effect of EPD Voltage on the Film Microstructure

The applied voltage can affect the macro and microstructure of the deposited film. Fig. 4 shows the SEM images of green

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Bi$_2$Te$_3$ film surface topography at two different EPD voltages of 100 V (a) and 50 V (b) for 10 minutes. It shows that by decreasing the voltage, the roughness of the surface decreases. However, in principal, this can lead to a reduction in the mechanical properties as well as the deposition weight and thickness of the Bi$_2$Te$_3$ films, which are desirable neither for handling the green films nor for the potential TE applications [8], [9], [11]. Therefore, in order to accomplish a high quality film, achieving an optimal voltage and time is essential for EPD [8].

Fig. 4 The SEM images of green Bi$_2$Te$_3$ films at two different EPD voltages, (a) 100V and (b) 50V

Although the results showed that the films deposited at 100V had a better interfacial adhesion to the substrate and higher deposition weight, there were small cracks in the film, and the surface was not smooth enough, as can be seen from Fig. 4 (a). Therefore, a voltage of 50 V is considered as the optimum deposition voltage for this suspension because it has deposited a film with higher quality.

IV. Conclusion

In summary, a crack-free and high quality p-type Bi$_2$Te$_3$ film has been successfully deposited by EPD for TE applications for the first time. A stable EPD suspension of p-type Bi$_2$Te$_3$ has been prepared in a mixture of acetone-ethanol with the use of triethanolamine as a stabilizer. The results have shown that the relationships between the deposition weight and EPD-related process parameters such as applied voltage and time were linear, which is all in good agreement with EPD theoretical principles. The optimum voltage and time of the EPD process was 50 V and 10 minutes, respectively, to achieve a high quality film on a copper substrate. An investigation of the morphology and microstructures of the green deposited films showed that a high quality, crack-free thick film of p-type Bi$_2$Te$_3$ has been achieved for TE applications.

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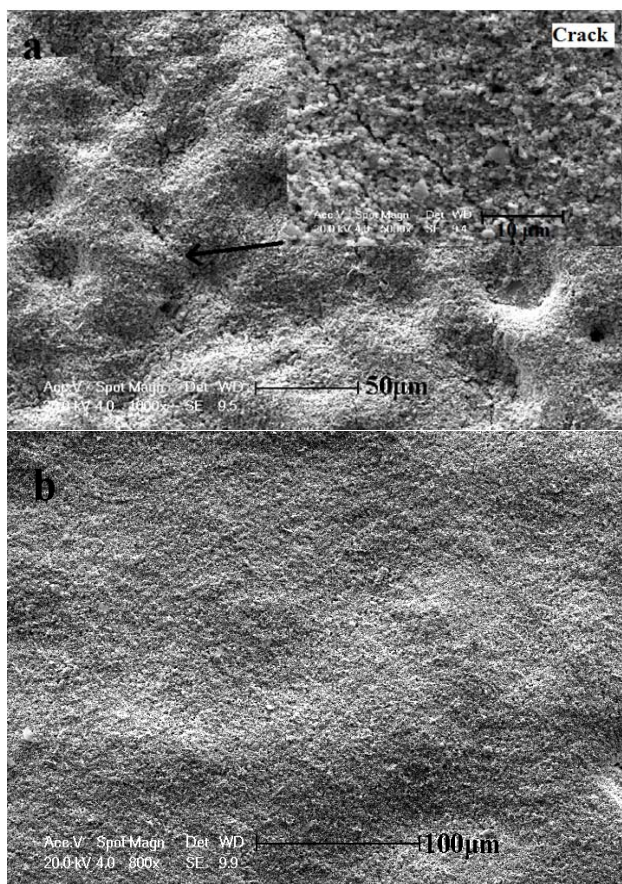
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