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Sequential Evaporation of Bi-Te Thin Films with Controllable Composition and Their Thermoelectric Transport Properties

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We report a simplified sequential evaporation route that can deposit compositionally controllable Bi-Te thermoelectric (TE) thin films without the need for a highly controlled facility. Te and Bi granules were used as starting materials, with their ratio being adjusted to obtain Bi-Te films with different compositions and thicknesses. The as-evaporated and annealed films were subjected to structural and morphological analysis, and their transport properties were measured. X-ray diffraction data revealed multiple phases for most films. Energy-dispersive x-ray spectroscopy showed that the film composition was Te-enriched due to the large vapor pressure difference of Te and Bi. A Bi_2Te_3 single phase was obtained in the annealed films, having nominal composition of $BiTe_{1.2}$. The existence of impurity phases, such as Bi_4Te_3 or elemental Te, was found in all the as-evaporated films and in the annealed films with other nominal Te/Bi ratios, which degraded the TE properties of the films by increasing their electrical conductivity and reducing their Seebeck coefficient. A pure Bi_2Te_3 film with nominal Te/Bi ratio of 1.2 exhibited a maximum power factor of 7.9×10^{-4} W m^{-1} K^2 after annealing at 200°C. This work demonstrated a simple, undemanding, reliable method to deposit Bi-Te-based TE thin films that can be utilized to fabricate low-cost TE microgenerators.

Key words: Bismuth telluride, thin film, sequential evaporation, annealing, transport properties

INTRODUCTION

During the last decade, thermoelectric (TE) materials and devices have been extensively studied due to their potential for power generation from existing temperature gradients in the environment and effective point cooling for low-scale and high-power electronics.[1–4] Thanks to advanced microfabrication and thin-film technologies, TE generators can be miniaturized, made more flexible, and integrated with sensors to autonomously power microelectromechanical systems (MEMS) devices in the future.[5–8] As the best room-temperature TE materials to date, with figures of merit near unity, Bi_2Te_3-based thin films are the most suitable candidate for microgenerators or microcoolers. Many reports have demonstrated the preparation of Bi-Te thin films, including by thermal evaporation,[9] sputtering,[10] pulsed laser deposition,[11] chemical vapor deposition,[12] and electrochemical deposition.[13]

Thermal evaporation is a well-developed technology and is capable of depositing various thin films with large area, high throughput, and low cost. For Bi-Te films, most reports highlight two different evaporation approaches, namely co-evaporation[6,9,14,15] and flash evaporation.[16–20] Regardless of the method used, control of film stoichiometry has always been a key factor which strongly affects the TE properties. For co-evaporation, composition

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