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Effect of the substrate on the electrodeposition of $Bi_2Te_{3-y}Se_y$ thin films

Luxia Bu [a], Wei Wang [a,*], Hui Wang [b]

[a] Department of Applied Chemistry, School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, PR China
[b] Analytical Center, Tianjin University, Tianjin 300072, PR China

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Abstract

The potentiostatic electrodeposition of n-type $Bi_2Te_{3-y}Se_y$ thermoelectric films onto stainless steel and gold substrates from nitric acid aqueous solutions has been carried out at room temperature. The cathodic process during the electrodeposition of $Bi_2Te_{3-y}Se_y$ films was investigated by cyclic voltammetric experiments. The structure and surface morphology of $Bi_2Te_{3-y}Se_y$ films deposited on both substrates were characterized by X-ray diffraction (XRD) and environment scanning electron microscopy (ESEM) coupled with energy dispersive spectroscopy (EDS). Electrical and thermoelectric properties of as-deposited films were also measured at room temperature. The results show that the reduction process under the same depositing conditions on gold and stainless steel substrates is very different. On gold substrates, H_2SeO_3 in the electrolyte is firstly reduced to elemental Se, and then the deposited Se reacts with $HTeO_2^+$ and Bi^{3+} to form $Bi_2Te_{3-y}Se_y$ alloy. On stainless steel substrates, $HTeO_2^+$ in the electrolyte is firstly replaced by elemental Fe to produce elemental Te, and subsequently the generated Te reacts with H_2SeO_3 and Bi^{3+} to form $Bi_2Te_{3-y}Se_y$ alloy. Analysis of ESEM show that the surface morphology of the films electrodeposited on gold substrates is more compact than that on stainless steel substrates. The XRD patterns indicate that the films electrodeposited on both substrates exhibit preferential orientation along (1 1 0) plane, but the relative peak intensity of (0 1 5) and (2 0 5) planes on stainless steel substrates is stronger than that on gold substrates. The Seebeck coefficient and electrical resistivity of the films deposited on stainless steel substrates are higher than that on gold substrates.
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1. Introduction

Bismuth telluride (Bi_2Te_3) and its derivative selenide compounds are considered to be the best materials for use in thermoelectric devices at room temperature [1]. Traditionally, bulk Bi_2Te_3 based materials have been prepared by the solid-state reactions at elevated temperatures [2–4]. However, an increasing interest is expected to develop with advances and discoveries in micro-thermoelectric devices [5]. Electrodeposition is one of the potential techniques to fabricate thin films for micro-systems. Compared with many other methods including chemical vapor deposition (CVD) [6,7], flash evaporation [8,9] and co-evaporation [10], electrodeposition offers the advantages of simple, easy operation and low cost. Moreover, the growth rate and composition of the films can also be easily controlled through

* Corresponding author. Tel.: +86 22 27402895; fax: +86 22 27409483.
E-mail address: wwangg@vip.sina.com (W. Wang).

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adjusting the electrodepositing parameters. Recently, several studies on the electrodeposition of Bi_2Te_3 thin films onto Pt, Mo, Ni, Ti and stainless steel, have been studied by many researchers [11–16]. Michel et al. have prepared $Bi_2Te_{2.7}Se_{0.3}$ thin films on stainless steel substrates by the chemical deposition method [5]. The formation of $Bi_2Te_{3-y}Se_y$ films has been claimed, in general, but no discussion on the possible influence of the substrate on the properties of the electrodeposit has been brought about. However, it is well known that the nature of the substrate plays a very important role on the initial formation of any electrodeposit, with consequences on their morphological and structural properties.

The choice of stainless steel substrates is due to a typical support for use in preparation of thermoelectric films [5,15]. Au can be used as a substrate to fabricate thermoelectric micro-generator [17], presenting interesting properties, such as good adhesion for electrodeposition and high overpotential for hydrogen evolution in acid aqueous solutions. In this paper, we report a comparative study on the characteristics of the electrodeposited $Bi_2Te_{3-y}Se_y$ films onto stainless steel and gold substrates. The effect of the substrate on the morphology, structure and thermoelectric performance of the electrodeposited $Bi_2Te_{3-y}Se_y$ films was investigated in details.

2. Experimental

2.1. Substrates

Gold and stainless steel sheets with geometric area of approximately 0.64 cm^2 were used as the substrates for the electrodeposition of $Bi_2Te_{3-y}Se_y$ films. In order to get good adhesion between the electrodeposited films and the substrates, a special attention was paid to the pretreatment of the substrates. The substrates were mechanically polished to mirror surface, and then degreased by acetone, etched in concentrated HNO_3 later, and rinsed with redistilled water at last. The pretreatments were performed just before the substrates were immersed into the solution to begin electrodeposition or cyclic voltammetry measurements.

2.2. Deposition conditions

Concentrated nitric acid was used to dissolve $Bi(NO_3)_3{\cdot}5H_2O$, H_2TeO_3 and H_2SeO_3, and the solution was subsequently diluted to 1 M HNO_3. The concentration of Bi^{3+}, $HTeO_2^+$ and H_2SeO_3 in the solution was 10, 9.5 and 3 mM, respectively. $Bi_2Te_{3-y}Se_y$ films were prepared by potentiostatic electrodeposition at room temperature using a conventional three-electrode cell consisting of gold and stainless steel sheets as working electrode, Pt plate as the auxiliary electrode and saturated calomel electrode (SCE) as the reference electrode.

CHI660B electrochemical working station was used to keep the deposition potential. Cyclic voltammograms were recorded with CHI660B electrochemical station at scan rate of 10 mV/s without stirring. All potentials were measured and expressed relative to SCE.

2.3. Characterization of the electrodeposited films

The morphology and composition of the deposited films were studied using a environment scanning electron microscope (ESEM, PHILIPS XL30) equipped with energy dispersive spectroscopy (EDS, OXFORD ISIS300). The X-ray diffraction (XRD) patterns of the as-deposited $Bi_2Te_{3-y}Se_y$ thin films were investigated in the diffracting angle range 20–110° with a film X-ray diffractometer (X'Pert Pro) using Co Kα radiation (λ = 0.17903 nm). The Seebeck coefficient of the films was measured with the thermoelectric performance measurement system (TPMF-100) developed by Tianjin University. Electrical resistivity (ρ) was measured by the four-point probe method. And the Seebeck coefficient and electrical resistivity were measured along the direction parallel to the surface of the films at room temperature.

3. Results and discussion

3.1. Voltammetric studies

The purpose of the cyclic voltammetric study was to define, for each substrate, the potential region in which the reactions occur. The voltammetric curves (the first cycle) measured by gold and stainless steel substrates are shown in

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