



Electrodeposition of Zn(O,S) (zinc oxysulfide) thin films: Exploiting its thermodynamic and kinetic processes with incorporation of tartaric acid

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ABSTRACT

Zn(O,S) (zinc oxysulfide) is an important chalcogenide material recently reported to be potentially applied as electrode buffers in thin film solar cells. Both vacuum and solution approaches have enabled the fabrication of Zn(O,S) films. However they either require extreme conditions and high energy consumption for synthesis, or suffer from lack of controllability mainly due to the thermodynamic and kinetic distinction between ZnO and ZnS during film growth. Here we demonstrated an effective electrodeposition route to obtain high-quality Zn(O,S) thin films in a controllable manner. Importantly, tartaric acid was employed as a secondary complexing agent in the electrolyte to improve the film morphology, as well as to adjust other key properties such as composition and absorption. To elucidate the vital role that tartaric acid played, thermodynamic and kinetic processes of electrodeposition was investigated and discussed in detail. The accumulative contribution has shed light on further exploit of Zn(O,S) with tunable properties and optimization of the corresponding electrodeposition process, for the application in thin film solar cells.

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1. Introduction

Zn(O,S) (zinc oxysulfide), which denotes the solid solution of ZnS and ZnO, is a promising semiconductor material used in buffer layers in thin film solar cells. It has been intensively investigated in the applications of CIGS [1], CZTS [2,3], SnS [4,5] and Cu₂O [6] solar cells. Zn(O,S) is originally developed to replace the toxic CdS buffer layer, since it consists of earth-abundant and non-toxic elements only. Besides, it has a large and tunable bandgap of 2.6–3.6 eV [7,8] comparing to that of CdS (2.4 eV), favorable to deliver a wider transmittance in the range of visible light. In addition, Zn(O,S) could be adjusted to have a much smaller bandgap than that of ZnO (3.2 eV) and ZnS (3.6 eV). In thin film solar cells like CIGS or CZTS, it is expected to provide significant bandgap bowing effect [8,9] compared with pure ZnO and ZnS, when considering band offset between buffer layer and absorber layer.

Several techniques have been employed for deposition of Zn(O,S) thin films [1], including chemical bath deposition (CBD) [3,5,10–12], sputtering [8,13], atomic layer deposition (ALD)

[2,4,14] and pulsed laser deposition (PLD) [7]. There were also novel approaches like aerosol assisted CVD [33] or photochemical method [34]. While vacuum approaches would have accurate composition control and refined grain growth, they need extreme environment with considerable energy consumption, which is not preferred in scaling up for mass production. CBD, though cheap and simple, is not able to deposit the targeting thin films in a controllable manner, mainly due to the thermodynamic and kinetic distinction between ZnO and ZnS during film growth [15–18].

To mitigate the drawbacks of these methods, electrodeposition would be a potentially alternate approach for low-cost, high-quality fabrication. Electrodeposition is a powerful technique to deposit metal as well as chalcogenide [35,36]. It demands no vacuum condition so both energy consumption and procedure complexity can be reduced compared to vacuum methods, while more precise control than CBD could be achieved by carefully tuning the deposition potential and current. Fathy and Ichimura first proved the feasibility of electrodepositing Zn(O,S) by a three-step pulse method [37]. To achieve better film quality and deposition controllability, we used a modified approach of electrodeposition by introducing tartaric acid (TA) as an additional complexing agent in this work. High quality, mirror-like Zn(O,S) thin films were obtained with the addition of tartaric acid and optimized depositing parameters. Morphology, composition, dynamic and

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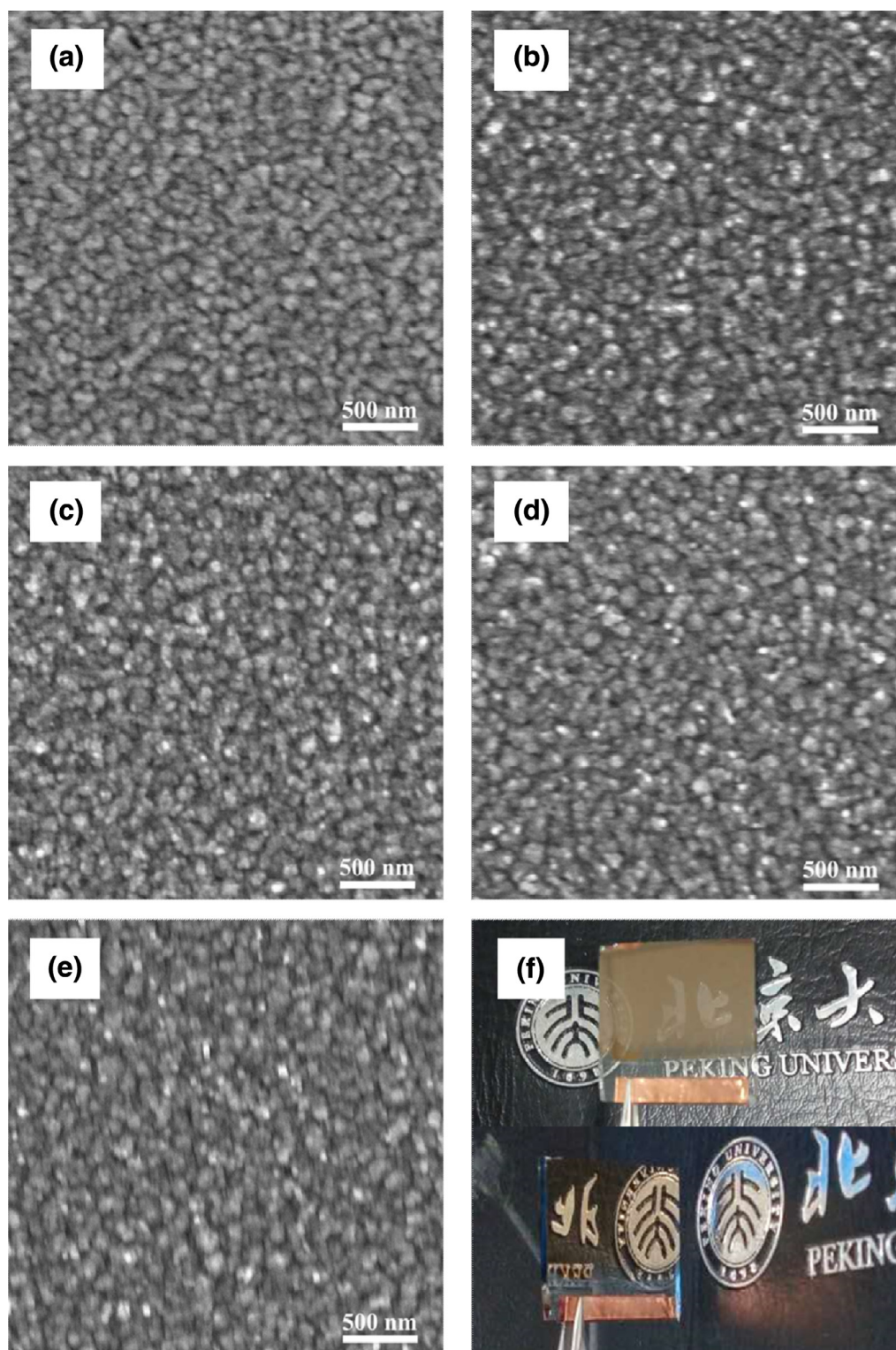


Fig. 1. (a–e) SEM top-view images of thin films deposited from the standard electrolyte with (a) 0 mM, (b) 25 mM, (c) 50 mM, (d) 75 mM, (e) 100 mM tartaric acid, respectively. The accelerating voltage was 10 kV and emission current was 10 μ A. (f) Optical images of deposition product (c).

optical properties were characterized subsequently. It showed that electrodeposition was an effective method to obtain high quality Zn(O,S) films, and the incorporation of tartaric acid greatly enhanced the quality and controllability of the as-prepared product. Moreover, thermodynamics and kinetics of Zn(O,S) electrodeposition were discussed in detail to elucidate the key role that tartaric acid played.

2. Experimental

Zinc oxysulfide thin films were deposited by potential static electrodeposition method using an electrochemical station (CHI660E, Shanghai CH Instruments, China). The deposition was carried out with a conventional three-electrode system, with FTO-coated glass (working electrode, active area $2.5 \text{ cm} \times 2 \text{ cm}$), Pt

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