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Comparison of the properties of CZTS semiconductor films grown by sequential and single step electrodeposition techniques

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Cu₂ZnSnS₄ (CZTS) is a promising semiconductor material suitable for application in low-cost and environmentally friendly thin film solar cells due to its superior optoelectronics properties. It is a perfect absorber material due to its high absorption coefficient ($>10^4 \text{ cm}^{-1}$) and direct optical bandgap (1.4-1.5 eV). Among the CZTS preparation techniques, electrodeposition is an attractive technique because of its simplicity, low cost and easy process controlling capability. In this investigation, a comparative study on CZTS films grown by two different techniques, namely, sequential electrodeposition and single step electrodeposition, has been carried out. Electrodeposition of Cu, Sn and Zn stack layers followed by sulphurisation with H₂S is one of CZTS growth techniques. In this study, growth parameters of sequentially electrodeposited CZTS were optimized to obtain best photoactive CZTS thin films. Electrodeposition parameters of Cu, Sn and Zn have been obtained using voltammograms. Cu thin film was electrodeposited on Mo substrate at -0.89 V vs Ag/AgCl in an electrochemical cell containing 0.4 M CuSO₄, 3 M lactic acid and NaOH at pH 11. Deposition of Sn thin film on Mo/Cu electrodes was carried out at -1.2 V vs Ag/AgCl in an electrochemical cell containing 0.055 M, 2.25 M NaOH and 8 ml of sorbitol. Zn thin film was electrodeposited on Mo/Cu/Sn at -1.2 V vs Ag/AgCl in an electrochemical cell containing 0.2 M ZnSO₄. In order to grow CZTS, Mo/Cu/Sn/Zn thin films were annealed at 550 °C for 60 min in H₂S. In the single step electrodeposition, CZTS thin films on Mo substrate were potentiostatically electrodeposited at -1.05 V vs Ag/AgCl for 40 min in a three electrode electrochemical cell containing 0.02 M copper (II) sulfate pentahydrate (CuSO₄·5H₂O), 0.01 M zinc sulfate heptahydrate (ZnSO₄·7H₂O), 0.02 M tin sulfate (SnSO₄) and 0.02 M sodium thiosulfate (Na₂S₂O₃) at room temperature. 0.2 M tri-sodium citrate (C₆H₅Na₃O₇) was used as the complexing agent and tartaric acid (C₄H₆O₆) was used as the pH control solution. The pH of the bath was maintained at 5. The Ag/AgCl and platinum electrodes were used as the reference and the counter electrodes respectively. Then samples prepared were annealed at 550 °C for 30 min in H₂S. CZTS films grown by two techniques were characterized using X-ray diffraction, reflectance, dark and light I-V, spectral response and C-V measurements in a PEC containing 0.1 M sodium acetate. Reflectance measurements reveal that the band gap energy of the films is 1.45 eV and I-V and spectral response measurements reveal that CZTS thin films were photoactive and p-type. The results obtained revealed that high quality photoactive CZTS can be prepared using both techniques. However, I-V and spectral response characteristics revealed that photoactive properties of CZTS thin films prepared by single step electrodeposition technique are superior in comparison to sequentially electrodeposited thin films.

Keywords: Cu₂ZnSnS₄, Cu/Sn/Zn metal stack layers, I-V characteristics, Single step electrodeposition

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