Tailoring Cu_2CdSnS_4 Phase Evolution for High-Efficiency Solar Cells via Precursor Engineering

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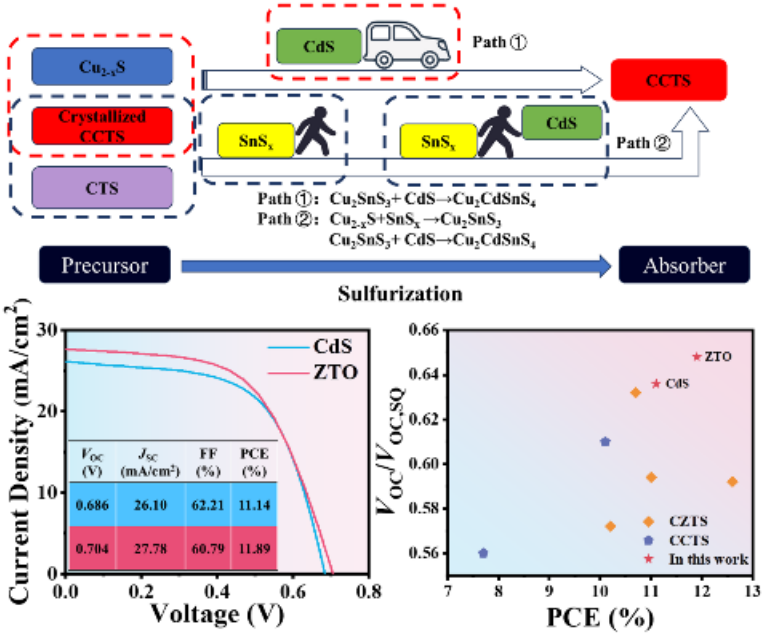
Pure sulfide stannite-based Cu_2CdSnS_4 (CCTS) solar cells, with substitution of Cd with Zn in Cu_2ZnSnS_4 (CZTS) solar cells, appear as potential alternations due to the similar photovoltaic properties to CZTS but the significant modification of V_{OC} deficit. CCTS solar cells demonstrate the merits as reduced Cu-Cd cation disorder due to the larger ionic radius of Cd2+ compared to Zn2+, and a higher formation energy for deep-level defects and defect clusters ($[2Cu_{Cd}^{+}+Sn_{Cd}^{2-}]$ vs. $[2Cu_{Zn}^{+}+Sn_{Zn}^{2-}]$). The materials have been achieved an impressive power conversion efficiency of 10.14% but lag behind CZTS, where the main limitation lies in the vague, complicate and uncontrollable phase evolution during the sulfurization leading to poor absorber quality. Herein, we propose an in-depth investigation of the effects of anions on alcoholysis and condensation reactions during the spin-coating process, as well as their nucleophilicity-mediated influence on impurity ion removal, leading to the different formation of intermediate phases (e.g. $Cu_{2-x}S$ or Cu_2SnS_3) in precursor films thereby altering phase transformation pathways during the subsequent sulfurization process. Ultimately, an impressive efficiency of 11.89% was obtained for air-solution-processed CCTS solar cells, highlighting significant research progress compared to CZTS and the highest stannite-based solar cells. Furthermore, a noteworthy $V_{OC}/V_{OC,\,SQ}$ of 65.0% was simultaneously obtained, exhibiting a comparable V_{OC}-deficit with high-efficiency CZTSSe and a leading position among the CZTS.



[1] J. Lin, Z. Huang, J. Zhao, S. Chen, H. Ma, L. Calvez, X. Zhang, C. Yan, Z. Su, G. Liang, *Adv. Funct. Mater.* 2025, 2509104.