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Formulation of Hybrid Nanopowder-Molecular Ink for Fabricating Critical Material-Free Cu_2ZnSnS_4 Thin Film Solar Absorber

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Abstract : Cu_2ZnSnS_4 (CZTS) compound (mineral name kesterite) has attracted considerable interests for photovoltaic application owing to its optoelectrical properties. Moreover, its elemental abundance in Earth's crust offers a comparative advantage for envisaged large-scale photovoltaic deployment without any material shortage issues. In this contribution, we present an innovative route to prepare CZTS solar absorber layer for photovoltaic application from low-cost and up-scalable process. CZTS layers were spin coated on the Molybdenum-coated glass from two inks composed of different solvents; dimethylsulfoxide (DMSO) and ultrapure water. Into each solvent; 0.57M $CuCl_2$, 0.39M $ZnCl_2$, 0.53M $SnCl_2$, and 1.85M Thiourea or $Na_2S_2O_3$, as well as pre-synthesized CZTS nanopowder, were added as sources of Cu, Zn, Sn and S in the ink. The crystallisation of ink into CZTS dense layers was carried out by firstly annealing the as-deposited CZTS layer in open air at 300°C for 1 minute, followed by sulfurisation at 560–620°C under atmospheric pressure for 120 minutes. Complementary electron microscopy, grazing incidence X-ray diffraction and Raman spectroscopy investigations suggest that both solvents can be used for preparing high quality and device relevant CZTS solar absorber layers. The sulphurisation crystallizes the as-deposited CZTS into highly polycrystalline CZTS layer with tetragonal structure demonstrated by the presence of tetrahedrally-shaped grains with the size of 1 µm. An advancement of the CZTS layer preparation was made by gradual substitution of volatile organic compound solvent of DMSO with ultrapure water. It is revealed that by using similar air annealing and sulphurisation process, dense and compact CZTS layers can also be fabricated from an ink with reduced volatile organic compound content.

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