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Solution-Processed Nontoxic and Abundant Cu_2ZnSnS_4 for Thin-Film Solar Cells

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Copper zinc tin sulfide (Cu_2ZnSnS_4, CZTS) is a very promising material as a low cost absorber alternative to other chalcopyrite-type semiconductors based on Ga or In because of the abundant and economical elements. In addition, CZTS has a band-gap energy of 1.4~1.5eV and large absorption coefficient over ~$10^4 cm^{-1}$, which is similar to those of Cu(In,Ga)Se_2(CIGS) regarded as one of the most successful absorber materials for high efficient solar cell. Most previous works on the fabrication of CZTS thin films were based on the vacuum deposition such as thermal evaporation and RF magnetron sputtering. Although the vacuum deposition has been widely adopted, it is quite expensive and complicated. In this regard, the solution processes such as sol-gel method, nanocrystal dispersion and hybrid slurry method have been developed for easy and cost-effective fabrication of CZTS film. Among these methods, the hybrid slurry method is favorable to make high crystalline and dense absorber layer. However, this method has the demerit using the toxic and explosive hydrazine solvent, which has severe limitation for common use. With these considerations, it is highly desirable to develop a robust, easily scalable and relatively safe solution-based process for the fabrication of a high quality CZTS absorber layer.

Here, we demonstrate the fabrication of a high quality CZTS absorber layer with a thickness of 1.5~2.0 μm and micrometer-scaled grains using two different non-vacuum approaches. The first solution-processing approach includes air-stable non-toxic solvent-based inks in which the commercially available precursor nanoparticles are dispersed in ethanol. Our readily achievable air-stable precursor ink, without the involvement of complex particle synthesis, high toxic solvents, or organic additives, facilitates a convenient method to fabricate a high quality CZTS absorber layer with uniform surface composition and across the film depth when annealed at 530 °C. The conversion efficiency and fill factor for the non-toxic ink based solar cells are 5.14 % and 52.8 %, respectively. The other method is based on the nanocrystal dispersions that are a key ingredient in the deposition of thermally annealed absorber layers. We report a facile synthetic method to produce phase-pure CZTS nanocrystals capped with less toxic and more easily removable ligands. The resulting CZTS nanoparticle dispersion enables us to fabricate uniform, crack-free absorber layer onto Mo-coated soda-lime glass at 500 ^{o}C, which exhibits a robust and reproducible photovoltaic response. Our simple and less-toxic approach for the fabrication of CZTS layer, reported here, will be the first step in realizing the low-cost solution-processed CZTS solar cell with high efficiency.

Keywords: Thin film solar cells, solution processing, CZTS