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Engberg, Sara Lena Josefin; Lam, Yeng Ming; Schou, Jørgen

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Optimized Packing Density of Large CZTS Nanoparticles Synthesized by Hot-injection for Thin Film Solar Cells

Sara Engberg^{(1)*}, Yeng Ming Lam⁽²⁾, Jørgen Schou⁽¹⁾

⁽¹⁾DTU Fotonik, Technical University of Denmark, DK-4000 Roskilde, Denmark

⁽²⁾School of Materials Science and Engineering, Nanyang Technological University, Singapore

* Corresponding author. Email: sleen@fotonik.dtu.dk

The absorbing kesterite material, $\text{Cu}_2\text{ZnSn}(\text{S}_x\text{Se}_{1-x})_4$ (CZTS), is very promising for future thin film solar cells. The material is non-toxic, the elements abundant, and it has a high absorption coefficient. These properties make CZTS a potential candidate also for large-scale applications. Here, solution processing allows for comparatively fast and inexpensive fabrication, and also holds the record efficiency in the kesterite family. Unfortunately, the record cell is deposited with a highly toxic solvent, hydrazine. This toxic solvent can be avoided through the nanocrystal ink approach, but to maintain good control of the nanocrystal formation during the synthesis, it is necessary to have organic ligands on the surface of the particles. These ligands are often long alkyl chains that potentially limit the quality of the film and degrade its electronic properties.

For nanocrystal solution processing to be a feasible fabrication route in the future, the amount of carbon in the film has to be limited. Today, several methods are employed in order to surpass this barrier, for example ligand exchange. A successful ligand exchange was carried out by Carrete *et al.* [1], where they replace the organic ligands by an antimony salt; however the efficiency is 1.4% for a cell annealed in Se-atmosphere.

In our work, we try to limit the carbon amount in the film by synthesizing larger nanoparticles. The bigger the particles are the smaller surface-to-volume ratio they have, which might decrease the amount of ligands necessary to stabilize the particles in solution. Today, CZTS nanoparticles synthesized through the so-called hot-injection method vary between 2 nm and 60 nm in diameter. In our group, we have synthesized particles larger than 200 nm. Transmission electron microscopy (TEM) allows us to image the faceted/hexagonal nanoparticles and determine their individual composition.

Densification of the film will also improve the film-quality. The optimal packing density will be calculated, and size-selective methods can be carried out in order to try to isolate the desired particle sizes. Films will be deposited through wet-chemical means, e.g. doctor-blading, spin-coating and spray-coating. The annealing time required can be minimized when starting with larger nanoparticles, and thus the elemental losses associated with annealing at higher temperature reduced. The films are characterized by TEM and scanning electron microscopy (SEM) as well as other surface characterization techniques.

A photovoltaic device of the structure soda lime glass (SLG)/Mo/CZTS/CdS/ZnO is built, and the power conversion efficiency will be determined. Our first CZTS solar cell made from doctor blading of approx. 20 nm $\text{Cu}_2\text{ZnSnS}_4$ nanoparticles in octanethiol, annealed in Se-atmosphere, had an efficiency of 1.4%.