Deposition and Characterization of Solution-Processed Chalcogenides for Photovoltaic Applications

Thesis Abstract

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Combating climate change requires society to shift to using clean, renewable sources of energy as quickly as possible. Photovoltaics (PVs) are a promising source of renewable energy due to the broad availability of solar radiation over the Earth's surface and the low cost of PV modules. While silicon solar cells dominate the current PV market, some drawbacks motivate the search for other solar materials. Silicon's indirect band gap necessitates using thick (>100 μm) absorber layers which limits applications to rigid substrates, and manufacturing silicon wafers suitable for solar cell applications requires slow batch processes, hindering the rapid deployment of PV technology.

One opportunity for realizing rapid manufacturing of PV modules is solution processing, wherein a solar cell is deposited with the use of liquid solutions containing the necessary constituent elements. A solution processing approach could be done in a roll-to-roll format in which a flexible substrate is coated at high speed to create a thin, flexible PV device. Such an approach is expected to dramatically increase the throughput capability of a photovoltaic manufacturing line. To realize the benefits of solution processing, suitable liquid-phase chemistries must be developed to enable the deposition of the desired absorber material while minimizing the incorporation of undesirable contaminants. One such approach is the amine-thiol solvent system which is notable for its ability to solubilize not only metal salts, but also metal sulfides, metal selenides, and pure metals. This makes the amine-thiol system a promising candidate for the deposition of metal chalcogenide absorber layer materials.

In this work, the chemistry of the amine-thiol system is studied in detail and reaction mechanisms governing the interaction of amine-thiol solutions with precursors relevant to the $Cu(In,Ga)(S,Se)_2$ material system are investigated. Nuclear Magnetic Resonance, Mass Spectrometry, and X-Ray Absorption measurements are performed to study this system. Structures for the metal thiolate species that form in these reactions are proposed, along with the products of the pyrolysis reaction that converts the thiolate species to the desired metal sulfides. The utility of this understanding is discussed.

The amine-thiol system is further applied to the synthesis of $AgIn(S,Se)_2$, a material with some similarities to the more common metal chalcogenide $CuInSe_2$ but studied far less thoroughly. The material and optoelectronic properties of $AgIn(S,Se)_2$ are characterized. X-Ray Diffraction, Hall Effect Measurements, Kelvin Probe Force Miscropscopy, and Quantitative Photoluminescence are all performed on $AgIn(S,Se)_2$ thin films. $AgIn(S,Se)_2$ films are found to exhibit high carrier mobility, benign grain boundaries, and strong photoluminescence emission,

suggesting that $AgIn(S,Se)_2$ may function as an effective absorber layer material for thin-film solar cells. Challenges facing its successful adoption as a solar cell material are discussed.

In this work, a novel method is developed to calibrate photoluminescence equipment for absolute photon counts, enabling one to calculate the absolute number of photons leaving a photoluminescence sample. This enables an estimation of the Quasi-Fermi Level Splitting of an absorber layer (and hence the open-circuit voltage of a solar cell) while only measuring a bare absorber layer film. The experimental method and required numerical analysis of the data are described herein.