ABSTRACT

McLeod, Steven M. Ph.D., Purdue University, August 2017. Selenization and Grain Growth of Thin-Film CIGSSe from Nanoparticle Inks. Major Professor: Rakesh Agrawal.

Humanity has become reliant on cheap, readily available energy to power our devices and maintain our modern quality of life. The current source of this energy is primarily fossil fuel, a nonrenewable energy source that will not be able to supply our growing energy demands indefinitely. A push to find new sources of energy to replace environmentally hazardous coal, natural gas, and oil has led to increased research into alternative energy sources like wind, tides, water, biofuel and geothermal heat. Still, the most abundant renewable energy source, the sun, remains largely untapped. Learning how to efficiently harness solar power figures to be critical in our pursuit of a clean and abundant energy source. There are numerous challenges if solar power is to become the primary source of grid power, mainly stemming from the fact that it is not continuously available and would have to be stored for times when power is not being generated. However the foremost concern is to find an efficient and cost effective way to convert solar power to electricity, so that a future where solar power is a competitive alternative to fossil fuels becomes realistic.

Thin film solar cell technologies such as $Cu(In_xGa_{1\text{-}x})(Se_yS_{1\text{-}y})_2$ (CIGSSe) and CdTe are currently two candidates to compete with crystalline silicon for share of the photovoltaic market. Despite crystalline silicon's superior efficiency and current domination of the solar cell market, thin film technologies remain attractive for their reduced material and manufacturing costs. The efficiency gap continues to close with CIGSSe laboratory efficiencies reaching 21.0% but these improvements have only been achieved for vacuum deposition processes. A more scalable approach through solution

processing offers a route to further reduce processing and capital costs, but solution based efficiencies need to improve beyond the current record of 17.3% in order to compete with vacuum based methods.

Towards that end, we have developed a solution processing method for synthesis and deposition of nanoparticle $Cu(In_xGa_{1-x})S_2$ (CIGS) inks, which are then annealed at high temperatures in the presence of selenium gas in a process known as selenization. The selenization process is extremely important in the formation of the CIGSSe absorber layer and essentially controls the final film morphology, material properties, and electrical properties. Despite its critical impact being widely acknowledged, the process remains largely a black box. This is even more true for solution processing, which could be viewed as a non-ideal case that introduces sources of contamination and other restrictions that further complicates the picture.

The primary thrust of the research in this thesis is to understand how processing parameters and precursor properties interact during selenization to determine the morphology and material properties of the resulting CIGSSe film. The goal is to understand the underlying mechanisms of the selenization process, thus "opening" the black box– or at least taking a peak inside. This is realized in the form of a nucleation and growth model for the selenization of our nanoparticle films. The second goal goes hand in hand with the first, and that is to apply this new understanding of the selenization step to optimize our CIGSSe fabrication process. Improved device efficiency is the ultimate goal, but there are also opportunities to pursue novel applications such as flexible devices, bifacial, and tandem solar cells.