Lead sulfide quantum dots and their application for solar cells

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Thesis submitted for the degree of

Doctor of Philosophy

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July 2016
This thesis is dedicated to my loving mom and dad Uma and Arun Shrestha, my sister Agya Shrestha and my wife Reshma Shakya.
Declaration

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Aabhash Shrestha

Date: 20 July 2016
Abstract

Quantum dot sensitized solar cells (QDSSCs) are interesting third generation solar cells that have potential to address the current energy related issues due to their low manufacturing cost, ease of fabrication as well as good performance. Quantum dots (QDs) offer several advantages such as size tunable band gaps across a wide range of energy levels, high molar extinction coefficients and enhanced stability. Among them, colloidal near infrared (NIR) QDs of lead sulfide (PbS) are attractive due to their narrow bulk bandgap, large exciton Bohr radii and the possibility of multiple exciton generation. Utilizing these QDs in solar cells with extendable IR absorption is promising. However, the progress of PbS QDSSCs is lacking due to the limited understanding regarding the synthesis and surface chemistry of QDs. The development of QDSSCs is also hindered by lack of proper counter electrode materials for the reduction of electrolytes. Hence, further developments in the synthesis and application of new materials for QDSSCs are necessary.

This PhD project focuses on the materials development for PbS QDSSCs such as PbS QD synthesis, surface ligand exchange of PbS QDs, and the development of new counter electrode materials. The following researches are included in this thesis:

1) A robust method to synthesize monodisperse lead sulfide (PbS) QDs is presented. PbS QDs with different sizes is produced by stepwise heating of the preformed seed QDs in the presence of excess oleic acid. A combination of "living" monomer addition and Ostwald ripening is identified as the mechanism for such QD growth processes.

2) The detailed synthesis mechanism of PbS QDs is investigated. Here, the various synthesis parameters influencing the nucleation and growth of PbS QDs are elucidated.
In addition, the detailed understanding of the synthesis mechanism is used to guide the synthesis of PbS QDs at ultra-small regime.

3) A versatile solution phase ligand exchange of PbS QDs in the presence of Pb-thiolate as the exchanging ligands is presented. The ligand exchange procedure better preserves the optical properties of PbS QDs and is applicable to a number of ligand/solvent systems.

4) The implementation of PbS QDs in QDSSCs is presented. The treatment of PbS QD photoelectrodes with cadmium salts is necessary to maintain the stability of PbS QDs in polysulfide based electrolytes. In addition, the number of cycles of CdS and ZnS treatment is optimized to achieve a photoconversion efficiency of 1.77%.

5) Finally, N-doped CN_x/CNT hetero-electrocatalyst materials using polydopamine is synthesized, which are explored as counter electrode materials for dye-sensitized solar cell (DSSC). These CN_x/CNTs material show excellent electrocatalytic activities towards the reduction of tri-iodide electrolytes with the optimized solar devices using CN_x/CNTs showing comparable performance (7.3 %) to reference Pt based devices (7.1 %).
List of publications

This doctoral thesis is prepared in “Publication” format according to the “specifications for Thesis (2016)” of the University of Adelaide. The thesis includes the following publications that have been published, submitted for publication, or ready for submission:

1) Aabhash Shrestha, Shizhang Qiao, Sheng Dai, “Near infrared (NIR) lead chalcogenide QDs- recent progress in their synthesis, post-synthesis ligand exchange and applications in solar cells” (To be submitted)


6) Aabhash Shrestha, Munkhbayar Batmunkh, Joseph G. Shizhang Qiao, Shapter, Sheng Dai, “Enhancing the stability of pre-synthesized PbS quantum dot sensitized solar cells in polysulfide electrolyte by treating with cadmium salts”, (To be submitted)

Some relevant components of the work have also been presented in the following conferences:

1) Aabhash Shrestha, Munkhbayar Batmunkh, Joe Shapter, Shizhang Qiao, Sheng Dai “Hybrid carbon nanomaterials for highly efficient dye sensitized solar cells (DSSCs)” MRS 2015, Fall Meeting, Boston, USA, 2015


3) Aabhash Shrestha, Shizhang Qiao, Sheng Dai, “Precursor and ligand influenced growth mechanism and living chain polymerization of post focused Lead Sulphide Quantum dots” SA Polymer and Biotechnology Symposium, Flinders University City Campus, Adelaide, Australia, 2013
Acknowledgements

First and foremost, I would like to express my deepest gratitude to Associate Professor Sheng Dai for being a truly unique and wonderful supervisor. I’m grateful for his excellent guidance, patience and providing an excellent research environment. He has been supportive from my early days of candidature and his guidance helped me complete this thesis. I would also like to thank my co-supervisor Professor Shizhang Qiao for his guidance and motivation.

I would like to express my deepest gratitude for all our lab members and colleagues who have made my PhD studies fruitful and enjoyable. Especially I would like to thank Associate Professor Bo Jin, Dr. Hu Zhang and Associate Professor Jingxiu Bi for their motivation and encouragement. I would also like to thank my colleagues Munkhbayar Batmunkh, Umar Azhar, Yusak Haranto, Steven Cui, Amir Mellati, Lei yuan Guo, Bingyang Zhang, Masoumeh Zagar, Mailin mission and others for being wonderful friends and helping me succeed. In addition, I would like to thank Associate Professor Tak Kee, Professor Nigel Spooner and Professor Joe Shapter for allowing me to use their lab facilities and incorporating me in their research teams.

My grateful thank also goes to the staff of Chemical Engineering and colleagues in the department. Especially, I would like to thank Associate Professor Zeyad Alawahabi for being a wonderful postgraduate coordinator and my teaching mentor. I would also like to thank Dr. Quihong Hu, Dr. Sanaz Orandi and Jason Peak for their technical support. I have thoroughly enjoyed knowing these wonderful people and have cherished their company during my PhD studies.
Last but not the least; I would like to thank all my family and friends for their wonderful support in my life. Especially, I would like to thank my mom and dad for being wonderful parents and their unconditional love. I’m truly blessed to have such wonderful and caring parents. I would also like to thank my lovely sister for supporting my every step in life. Finally, I would like to thank my lovely wife for her love, support and for being a true friend.
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