Final Project Report (2005-2014)

Hydrogen Generation Using Integrated Photovoltaic and Photoelectrochemical Cells

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Program Scope

In the pursuit of efficient PEC hydrogen generation, the use of tandem cells of photoelectrochemical (PEC) and photovoltaic (PV) cells is a promising route. While PEC cells of a single photoanode have been constructed and characterized from various materials (TiO_2, WO_3, and ZnO), the efficiency have been low due to the lack of electromotive force (EMF) generated in comparison to the conduction band of the metal oxides and the reduction couple H^+/H_2O. External application of a voltage generated by a photovoltaic cell coupled to a PEC cell to properly generate the EMF of 1.23 eV necessary for water splitting. Further improvement of oxide catalysts is to engineer them in such a way that they absorb the solar spectrum (visible light) more efficiently. This requires research into oxide nanomaterials that are doped with elements such as nitrogen and/or sensitized with small bandgap semiconductor quantum dots (QDs), such as CdSe and CdTe.

Our specific aims are:

- Creating photoelectrochemical devices with enhanced visible light absorption and improved PEC performance by combing two methods of bandgap engineering, nitrogen doping *and* sensitizing with CdSe quantum dots.
- Synthesis and study of one-dimensional (1D) nanomaterials with N-doping and quantum dots, including WO_3, TiO_2, ZnO, CdSe and CdTe.
- Developing photoelectrochemical devices based on self-assembled CdSe quantum dots and TiO_2 colloidal spheres without the use of small molecule dielectrics.
- Understanding the fundamental processes involved in PEC based on nanomaterials, including charge transfer and transport.

Final Report

In this entire project period from 2005-2014, we have made significant progress in developing novel nanostructures of metal oxides (MOs) for solar hydrogen generation based on photoelectrochemical (PEC). Materials investigated are focused on 1D and 0D MO nanostructures of TiO_2, WO_3, ZnO, and Fe_2O_3 in conjunction with quantum dot (QD) sensitization and chemical doping (N or H) to alter their electronic band structures for both visible light absorption and for facilitating interfacial charge transport. In addition, we have used plasmonic metal nanostructures to enhance the PEC performance by improving light absorption of QDs via enhanced scattering of the plamonic metal. Most importantly, we have discovered a multipronged strategy for improving PEC performance: using plasmonic metal nanostructure to enhance light absorption, QDs to improve charge transfer, and chemical doping to increase

charge transport in metal oxides for PEC. The combination is critical for overall high efficiency of PEC. This strategy is developed and demonstrated for the first time to our best knowledge.

Specially, several major achievements are highlighted below:

- Demonstration of using plasmonic nanostructures to enhance light absorption of semiconductor QDs, thereby improving photocurrent of QD-sensitized MO nanomaterials for PEC hydrogen geneartion from water splitting.
- Determination of the mechanism behind improved photocurrent in PEC of hydrogen-treated MOs in terms of increase oxygen vacancies and majority charge carrier density.
- Direct probe of bandgap states in hydrogen-treated MOs and their relation to optical properties and photocurrent generation.
- Demonstration of core/shell and composite nanostructures involving Fe2O3 for PEC applications.
- Investigation of exciton and charge carrier dynamics on the ultrafast time scales to gain insight on influence of various factors such as interfaces between electron donors and acceptors using ultrafast laser and X-ray techniques
- Synthesis and characterization of a number of semiconductor QDs and MO nanostructures for PEC applications.

Publications from the entire project period (80 total)

80. Ying-Chih Pu and Jn. Zhang, "Mechanisms Behind Plasmonic Enhancement of Photocurrent in Metal Oxides," Austin J. Nanomed. Nanotech. (invited editorial), in press, 2014.
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Personnel Associated with the Research in the entire project period

- **At UCSC**

  Staci Admas (Ph.D. student, current)
  Sarah Lindley (Ph.D. student, current)
  Ying-Chih Pu (Postdoctor, current)
  Abe Wolcott (Ph.D. graduate student, graduated June 2009)
  Jenny Hensel (Ph.D. graduate student, graduated June 2011)
  Bob Fitzmorris (Ph.D. graduate student, graduated June 2012)
  Damon Wheeler (Ph.D. graduate student, graduated June 2012)
  Jason Cooper (Ph.D. graduate student, graduated June 2012)
  Liping Liu (Ph.D. visiting graduate student from Tsinghua University, China)
  Jianying Shi (visiting professor from Zhong Shan University, China)
  Haixia Ma (visiting professor from Northwest University, China)
  Tzarara Luke-Lopez (visiting graduate student and postdtcor,from Mexico)
  Omed Muzaffery (undergraduate student, graduated in June 2009)
  Jordan Edberg (undergraduate student, graduated in June 2011)
  Dr. Jin Z. Zhang (PI)

- **At UGA**

  Wilson Smith (Ph.D. graduate student, graduated in 2009)
  George Larsen (Ph.D. graduate student, graduated in 2012)
  Whitney Ingram (Undergraduate student)
  Dr. Yiping Zhao (Co-PI)

Current and Pending Support

Jin Zhang, UCSC

Current:

1. Renaissance Health Service Cooperation and Its Associates, "Synthesis and Bioconjugation of Hollow Gold Nanospheres (HGNs)", $219,120, 2012-2015, (PI)
   No overlap with the current DOE grant.
2. UC-MEXUS. "Investigating the Charge Transport Properties on Solar Cells Sensitized with Quantum Dots and Metallic nanoparticles", $25,000, 2013-2015, (Co-PI)
   Small overlap with the current DOE proposal in terms of the use of quantum dots.

Pending:

1. NIH, "Probing Mechanisms of Heat Transport and Cell Death in Photothermal Ablation", $381,512, 07/01/15-06/30/18, (PI)
   No overlap with the current DOE grant.
2. NSF, "Rational Codoping as a Strategy to Enhance Blue Luminescence of Ag+-Doped Semiconductor Quantum Dots for White Light Generation", $381,837, 07/01/15-06/30/18, (PI)
   No overlap with the current DOE grant.
3. NSF, "SusChem: Understanding the Origin of Instability of Perovskite Quantum Dots from a Dynamic Perspective", $479,000, 07/01/15-06/30/18, (PI)
   No overlap with the current DOE grant.

Yiping Zhao, UGA

Current:

1. NSF, "Rationally Designed Three-Dimensional Nanostructures for Surface Enhanced Raman Spectroscopy", $329,704, 10/01/10 – 03/15/15, (PI)/
   No overlap with the current DOE grant
2. U.S. Department of Agriculture, "Technologies as Multiple Hurdles to Inactivate Shiga Toxin-Producing Escherichia coli and Viruses during Beef Processing and on Non-intact Beef", $4,990,000, 01/01/2011 – 12/31/2015, (co-PI, with Y. Hung, etc)
   No overlap with the current DOE grant
3. NSF, "SERS Based Micro-Sensor Arrays for Quantitative miRNAs Detection", $409,999, 09/01/2011 – 08/31/2015, (PI, with R. A. Tripp, and J. Driskell)
   No overlap with the current DOE grant
4. NSF, "NUE: UNITE - Undergraduate Nanotechnology Inquiry, Training, and Experimentation at the University of Georgia (UGA)", $200,000,1/1/2013-12/31/2014, (co-PI, with Leidong Mao, Jason Locklin, Bingqian Xu, and Zhengwei Pan)
   No overlap with the current DOE grant
5. NSF, "Collaborative Research: Kinetics of Autonomous Catalytic Nanomotors in Confined and Crowded Environments, $359,930, 06/15/2013 – 06/14/2016, (PI, with Rui Qiao)
   No overlap with the current DOE grant
6. NSF, "Template Based Fabrication of Three Dimensional Optical Metamaterials, $250,001, 08/01/2014 – 07/31/2017, (PI, with Bill Dennis)
   No overlap with the current DOE grant
7. NSF, "Nanomotors for thrombolytic therapy after stroke, $275,000, 03/01/2014 – 02/28/2016, (co-PI, with K. L. Jin, Bo Yang)

No overlap with the current DOE grant

Pending:

1. NSF, "Collaborative Research: Growth of High Quality Heterostructured Oxide Nanorods through Energetic Shadowing Self-Assembly", $203,760, 08/01/2015 – 07/31/2018, (PI, with Bo Yang and Dexian Ye)

No overlap with the current DOE grant

2. NSF, "Collaborative Research: Assembly, Characterization, and Modeling of Functional Multi-component Nanomotors", $ 225,780, 05/01/2014 – 04/30/2017, (PI, with Bo Yang)

No overlap with the current DOE grant

3. NSF, "Collaborative Research: Active Magnetic Nanomotors to Enhance Thrombolysis for Better Ischemic Stroke Treatment, $ 391,139, 05/01/2015 – 04/30/2018, (PI, with Bo Yang, Leidong Mao and Brian Condie)

No overlap with the current DOE grant

Cost Status

Fund has been completely spent.