II.G.15 Center on Nanostructuring for Efficient Energy Conversion (CNEEC)

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Objectives

CNEEC's mission is to understand how nanostructuring can enhance efficiency for solar energy conversion to produce hydrogen fuel and to solve fundamental cross-cutting problems.

The overarching goal is to increase conversion efficiency by manipulating materials at the nanometer scale. We develop advanced synthesis, fabrication and characterization methodologies to understand how nanostructuring can optimize light absorption through quantum and optical confinement and improve catalysis through theory-driven and bio-inspired design. Each is manipulated to improve performance and efficiency in solar energy conversion to hydrogen fuel for storage.

Our research helps understand and expand the scientific foundation of the underlying physical and chemical phenomena that can lead to break-out high-efficiency, cost-effective energy technologies. This multi-disciplinary approach is enabled by the Center structure that provides the intellectual environment and the facilities infrastructure critical to carry out the research projects. A team of CNEEC researchers assembled across disciplines and institutions (see Fig. 1) bring their complementary expertise to bear on these complex but fundamental issues that cut across not just conversion of sunlight to hydrogen fuel, but also many energy conversion and storage devices. To pursue its mission, CNEEC has organized its research activities in two interconnected projects:

- Project 1. Optical and quantum confinement for light absorption.
- Project 2. Atomic-scale engineering for catalysis.

Technical Barriers

The two projects collectively aim to tackle two primary technical barriers: (1) the efficient absorption of sunlight and (2) subsequent conversion it to stored energy in the form of hydrogen fuel. The project teams work closely together to integrate the best absorbers from Project 1 with the best catalysts from Project 2 and test their solar-to-chemical conversion efficiency.

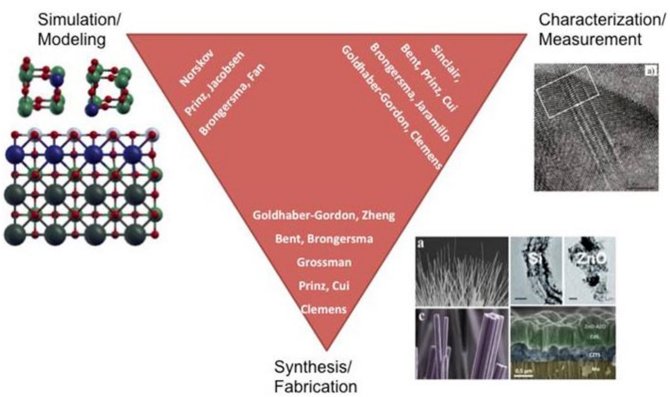

FIGURE 1. CNEEC model.

Abstract

This poster will cover selected CNEEC highlights from both Project #1 and Project #2, as well as their integrations, Fig. 2. In particular, the poster will describe our efforts to establish nanoconfinement effects, to utilize photonic concepts for enhanced light absorption, and to develop sophisticated fabrication and observation platforms to advance the field of photoelectrochemical (PEC) water-splitting. The poster will also describe our efforts in catalyst engineering at the atomic scale in order to develop active catalysts for both the hydrogen evolution reaction (HER) and the oxygen evolution reaction (OER). We will also show our latest results in developing active, stable, low-cost photoelectrodes for PEC water-splitting.

Progress Report

Selected key accomplishments in CNEEC during this past year:

- Demonstrated interparticle electronic coupling between closely spaced quantum dots using electron energy loss spectroscopy in the transmission electron microscope (STEM-EELS).
- Demonstrated the ability to engineer band energy positions of PbS quantum dots through passivation by ligands with different dipole moments.
- Used atomic layer deposition (ALD) to form engineered PbS quantum dots and Al_2O_3 barrier layers to improve charge collection of photo-induced carriers; tested using quantum dot-sensitized solar cell platform.

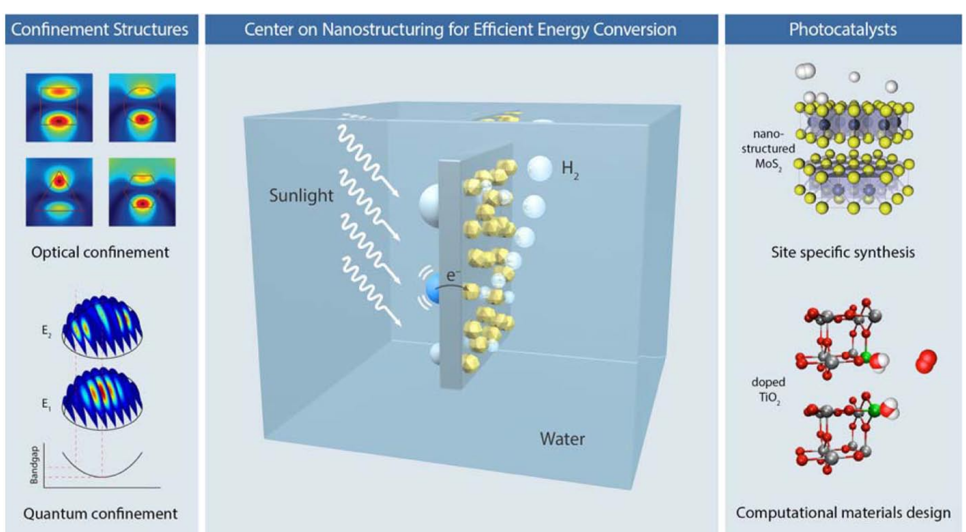

FIGURE 2. Schematic depiction of CNEEC research projects.

- Accomplished record absorption coefficients for visible light using self-assembled plasmonic arrays tuned by atomic layer deposition.
- Used optical simulations to demonstrate that judiciously engineered iron oxide photoanodes based on nanocone arrays can achieve total above-band-gap solar absorption.
- Developed new types of transparent electrodes consisting of mesoscale and nanoscale metal nanowires achieving a sheet resistance of 0.36 Ohm/sq and transmittance of 92%.
- Demonstrated large-area nanopatterned photoelectrodes that capitalize on optical (Mie) resonances to boost the rate of water splitting reactions by a factor of 3.
- Developed models for the performance limits on PEC water-splitting based on the current state of materials research, providing insights into avenues of greatest impact to improve performance.
- Identified the surface structure of manganese oxide catalysts under OER and ORR operating conditions using in-situ synchrotron spectroscopies.
- Engineered improved catalysts by interfacing manganese oxide with gold, and explored their interactions with *ex-situ* and *in-situ* synchrotron spectroscopies.
- Developed precious-metal free regenerative fuel cells for energy storage by means of water electrolysis, based on CNEEC-developed OER catalysts.
- Identified how MoS_2 surface structure impacts its semiconductor properties for PEC water-splitting.
- Employed theory to identify transition metal selenides for HER and to understand trends in reactivity based on the electronic structure.
- Developed methods to calculate Pourbaix diagrams to assess material stability under PEC water-splitting conditions, expanding screening-space to include layered perovskites, double perovskites, and 2400 additional known materials from the ICSD database, leading to the identification by theory of several new promising materials for visible light absorption and catalysis.
- Identified new promising catalyst materials by means of a computational DFT screening study of several hundred ABO_3 perovskite oxides, including strain-induced systems.
- Developed ternary oxide OER electrocatalysts deposited by ALD, complementing theoretical predictions on mixed metal oxide catalysts made by Nørskov and coworkers in CNEEC.
- Integrated atomically-engineered molybdenum sulfide catalysts onto silicon to produce highly active, and stable photocathodes for PEC water-splitting without precious metals.

Future Directions

CNEEC will continue forward with its mission is to understand how nanostructuring can enhance efficiency for solar energy conversion to produce hydrogen fuel and to solve fundamental cross-cutting problems. By manipulating materials at the nanometer scale through advanced synthesis, fabrication and characterization methodologies we will impact optical and catalytic properties of materials to produce fundamental advancements that can ultimately enable technology in this field.

Selected publications acknowledging the DOE grant or contract

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