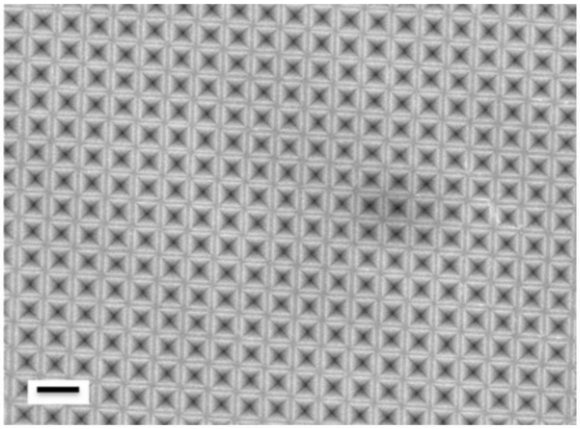
Energy: Photovoltaics, Energy Harvesting, Batteries, Fuel Cells

High-efficiency 10-µm-thick Thin Film c-Si Solar Cells Enabled by Inverted Nano-pyramid Light-trapping Structures

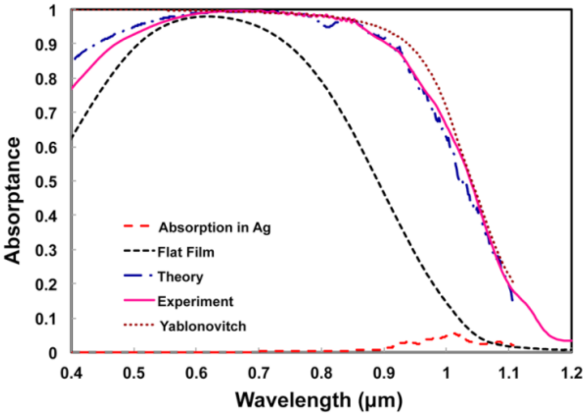
M.S. Branham, W. -C. Hsu, S. Yerci, G. Chen

Sponsorship: SunShot Initiative, Department of Energy, USA

Crystalline silicon (c-Si) is the dominant material in the photovoltaic industry, yet silicon is expensive and contributes ~40% to the total module cost of c-Si solar cells. Reducing the material intensity by creating thin-film devices is one strategy to reduce the overall cost of silicon PV. Here, we demonstrate experimentally that an inverted nanopyramid light-trapping scheme for a 10-µm-thick c-Si thin-film can achieve an absorptance value comparable to that of a 300-µm-thick planar device. Figure 1(a) shows a scanning electron microscope image of a 700-nm-pitch inverted nano-pyramids (INPs) light-trapping structure. In Figure 1(b), simulation shows the comparable ultimate efficiency with a 300-µm-thick planar device using a 10-µm-thick c-Si thin-film with INPs structure, and its corresponding reflection data are also measured in Figure 1(c). We are applying these light-trapping structure to thin silicon-on-insulator wafers to produce photovoltaic devices with current energy conversion efficiencies exceeding 13%. To reach the high efficiencies necessary for a commercial product, we also constructed a multi-physics optimization tool incorporating both optical absorption and an electronic carrier collection to understand in detail the loss mechanisms of the devices, including incomplete photonic absorption, contact recombination, surface recombination, and Schottky-Read-Hall and Auger recombination. Our model predicts that a 10-µm-thick thin-film c-Si solar cell with an inter-digitated back contact scheme can have an efficiency higher than 20%.



▲Figure 1: Scanning electron microscope image of inverted nano-pyramids (INPs) with a pitch of 700 nm. The scale bar is 1µm.



▲Figure 2: Comparison of theoretical and experimental absorptance spectra. Dimensions of the flat film structure are 70 nm SiNx, 10 µm Si, 250 nm SiO_2, and 200 nm Ag in thickness. The inverted pyramid structure consists of 90 nm SiNx, 10 µm Si, 0.5 µm SiO_2, and 200 nm Ag. Note that, except for Ag absorption curves, other absorptance spectra account for absorption in all layers.

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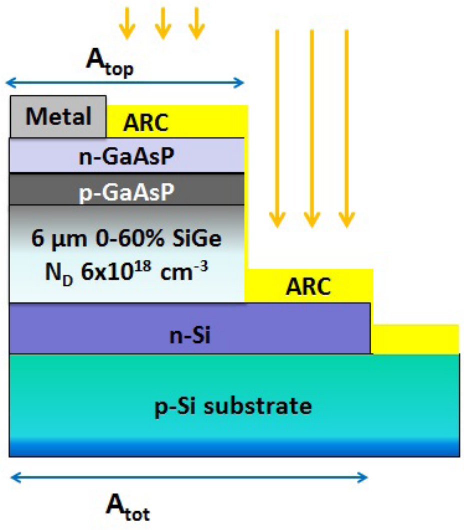
Step-cell Design for Tandem GaAsP/Si Solar Cells

E. Polyzoeva, S. Hadi, E.A. Fitzgerald, A. Nayfeh, J.L. Hoyt

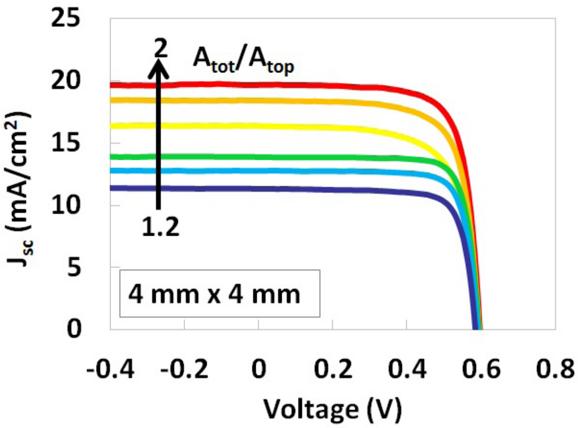
Sponsorship: Masdar Institute of Science and Technology

This work is part of a collaborative project aimed at ultimately achieving photovoltaic conversion with more than 40% efficiency at a lower cost by combining low-cost manufacturing on Si wafers with high-efficiency III-V materials in one tandem cell. In order to grow high quality GaAsP layers on the Si cell, a graded SiGe buffer is used to provide a transition from the lattice constant of the Si to that of the GaAsP, as in Figure 1. However, due to its smaller bandgap than Si, the SiGe buffer absorbs a portion of the light intended for the Si sub-cell and reduces the overall cell efficiency. In this work, we explore a step-cell design aimed at increasing the amount of light reaching the Si sub-cell.

Figure 1 shows the step-cell design for the tandem GaAsP/Si structure. Part of the GaAsP top cell and the SiGe graded buffer is etched away to expose the Si sub-cell. In the initial experiments, the step-cell design was implemented on a Si cell with SiGe layer grown on top to assess the effect of varying the area of exposed Si on the short circuit current of the solar cell. The current-voltage characteristics of the SiGe/Si cells as a function of the A_{tot}/A_{top} ratio are shown in Figure 2. The short circuit current increased from 11 to 20 mA/cm² when the ratio increased from 1.2 to 2. These results demonstrate that the step-cell design is effective in increasing the current of the Si sub-cell and therefore improving the overall efficiency. Going forward, the step-cell design will be implemented into the complete tandem structure, including the GaAsP sub-cell.



▲Figure 1: Step-cell design of GaAsP/Si tandem solar cell with SiGe graded buffer. Part of the GaAsP and SiGe layers is etched away to allow more light to reach the Si sub-cell and to provide higher overall efficiency.



▲Figure 2: J-V characteristics of a SiGe/Si step cell as a function of A_{tot}/A_{top} ratio. The structure is the same as in Figure 1 without the GaAsP layers. The short-circuit current increases from 11 to 20 mA/cm^2 when the ratio increases from 1.2 to 2.

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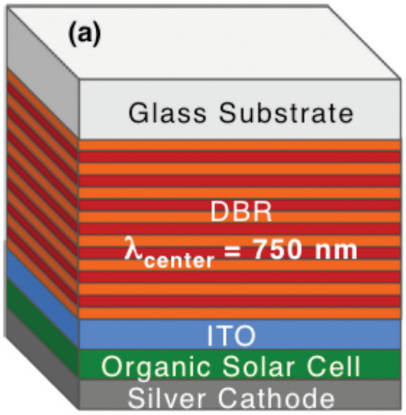
Slow Light-Enhanced Singlet Exciton Fission Solar Cells with 126% External Quantum Efficiency

D.N. Congreve, N.J. Thompson, D. Goldberg (CUNY), V.M. Menon (CUNY), M.A. Baldo
Sponsorship: Department of Energy, Basic Energy Sciences, DE-SC0001088

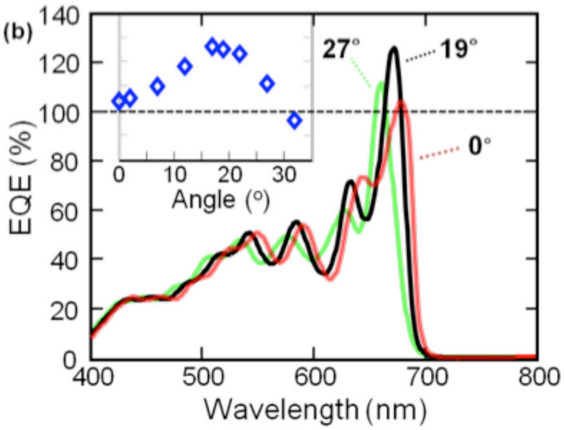
Singlet exciton fission can improve the electrical yield of solar cells without increasing the number of photovoltaic junctions by generating up to two electrons for every incident photon. A key measure of the efficiency of fission is the external quantum efficiency (EQE), the fraction of incident photons that are converted into electrons and delivered to the load. Recent demonstrations using pentacene have proven that singlet exciton fission in organic solar cells can deliver EQEs exceeding the benchmark 100%. The limiting factor in these devices is light absorption. Unfortunately, it is not possible to simply use a thicker layer of pentacene because its excitons decay before dissociating into charge.

Light management, however, is a feasible method to improve absorption within thin pentacene layers. We demonstrate a simple approach for enhancing absorption in thin film organic solar cells by exploiting the slow light modes that appear at the band edge of a distributed Bragg reflector (DBR). Using this approach we show over a 50% enhancement in absorption and EQE of singlet-exciton-fission-based solar cells.

When the DBR band-edge mode is tuned to the peak wavelength of pentacene's extinction coefficient, we observe an EQE peak of 126±1%, as in Figure 2. A control solar cell fabricated identically but without the DBR achieved a peak EQE of only 83% and exhibited nearly zero change in EQE with varying incident angle. The DBR-enhanced device demonstrated EQE greater than 100% for incident angles over the range +/- 27°, with a relatively flat response; see inset of Figure 2. This technology can greatly improve absorption without adding significant device complexity.



▲Figure 1: Device structure of the organic photovoltaic. A microcavity is created with the DBR and the silver cathode, boosting absorption in the organic layers.



▲Figure 2: External quantum efficiency as a function of incident light angle. The device shows a photocurrent enhancement for a wide angle range.

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Energy Level Modification in Lead Sulfide Quantum Dot Photovoltaics Through Ligand Exchange

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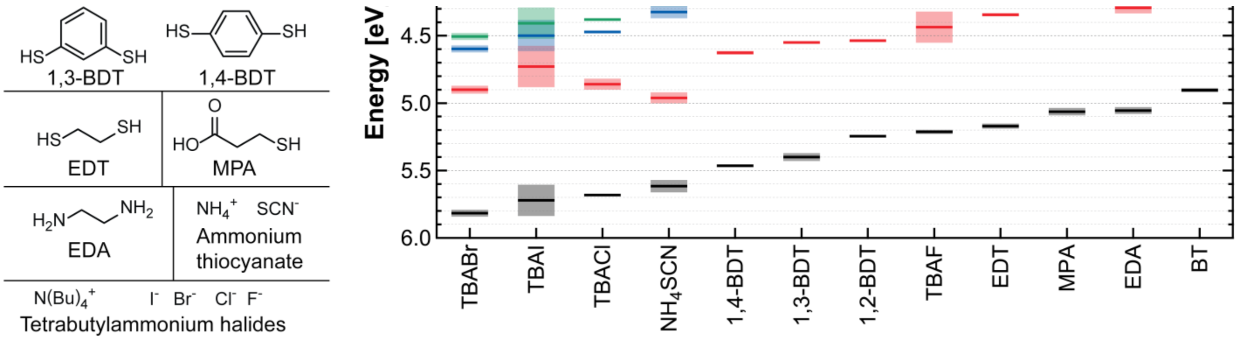
Sponsorship: Hertz Foundation, National Science Foundation, Samsung

The electronic properties of lead sulfide (PbS) colloidal quantum dots (QDs) are highly dependent on QD size and surface chemistry. Novel surface passivation techniques involving organic or inorganic ligands have contributed to a rapid rise in the efficiency of QD photovoltaics, yet the influence of ligand-induced surface dipoles on PbS QD energy levels and photovoltaic device operation is not yet fully understood. Ligand exchange treatment is known to shift the valence and conduction band energies of CdSe and InAs QDs, but the incidence of similar shifts in PbS QDs and their relevance to the operation of PbS QD photovoltaics have yet to be explored. Here, the valence band energies of PbS QDs treated with twelve different ligands are measured using ultraviolet photoelectron spectroscopy (UPS) and correlated with the results of atomistic simulations and photovoltaic device characterization. As shown in Figure 1, a valence band shift of up to 0.9 eV is observed between different ligand treatments. Treatments with 1,2-benzenedithiol and 1,3-benzendithiol, which result in valence band energies differing by ~0.2 eV, are employed for PbS QDs in three different solar cell architectures, and changes in device performance are correlated with the measured energy level shift. Atomistic simulations of ligand binding to pristine PbS(100) and PbS(111) slabs qualitatively reproduce the measured energy level shifts. These findings complement the known bandgap-tunability of colloidal QDs and demonstrate an additional level of control over the electronic properties of PbS QDs.



▲ Figure 1: Ligand-induced band energy shifts in PbS QDs. (a) Chemical structure of ligands studied here, including thiols (1,4-benzenedithiol (1,4-BDT), 1,3-benzenedithiol (1,3-BDT), 1,2-benzenedithiol (1,2-BDT), benzenethiol (BT), 1,2-ethanedithiol (EDT), and 3-mercaptopropionic acid (MPA)), primary amines (1,2-ethanediamine (EDA)), ammonium thiocyanate (NH4SCN), and tetrabutylammonium halides (iodide (TBAI), bromide (TBAB), chloride (TBAC), and fluoride (TBAF)). (b) Energy levels of ligand-exchanged 3.3-nm diameter PbS QDs measured by UPS (valence band and Fermi level) and absorption spectrophotometry (conduction band). The measured direction of the shifts correlates with computational simulations of ligands binding to PbS(100) and PbS(111) surfaces and with the performance of PbS QD photovoltaic devices.

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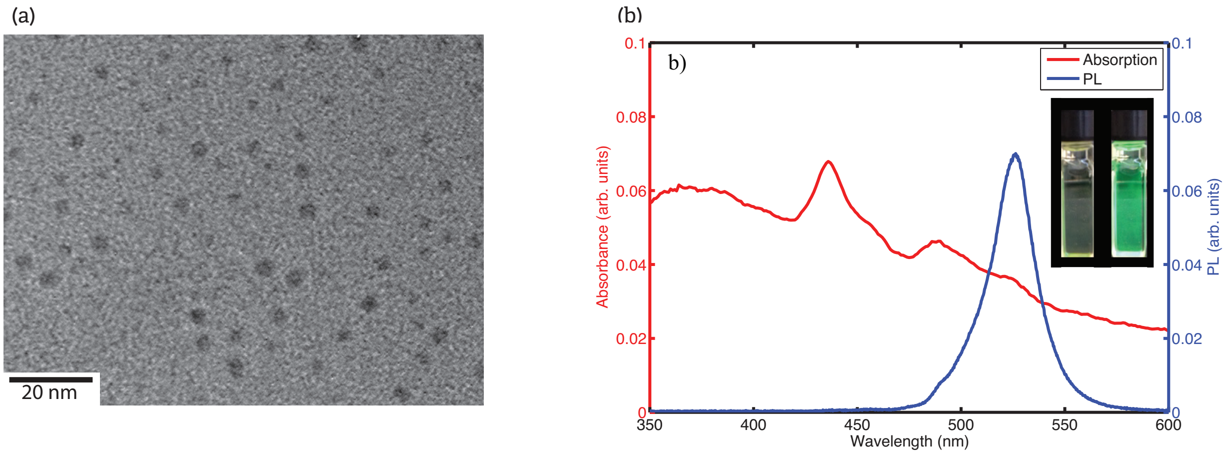
Optical and Structural Properties of Organohalide Perovskite Nanocrystals

P. Tyagi, W.A. Tisdale
Sponsor: U.S. Department of Energy

Over the last few years, organohalide perovskites have emerged as the most promising contenders in the field of photovoltaics. These materials are cost-effective, exhibit large carrier diffusion lengths (~1 micron), and have high power conversion efficiencies (> 14%). Although the general perovskite structure has been known for more than six decades, the unique composition of organohalide perovskites has shown favorable properties for photovoltaic applications. Most research on organohalides has focused on studying charge transport processes in thin films for device fabrication purposes. However, the properties of organohalides in the nanoscale length regime are mostly unexplored. We are currently investigating the correlation between structural and transport properties of perovskite nanocrystals. In particular, we have synthesized nanocrystals (NCs) of methylammonium lead bromide perovskites following the procedure of Schmidt et al. below. The NCs are nearly 4 nm in diameter (see Figure 1a) and exhibit bright emission in the visible (see Figure 1b). Experiments are underway in our lab to study transport properties of these materials using spatially and temporally resolved photoluminescence spectroscopy.



▲ Figure 1: (a) TEM, (b) absorbance (red), and photoluminescence (blue) of methylammonium lead bromide NCs. The inset of (b) shows a cuvette of NCs in the absence (left) and presence (right) of UV excitation.

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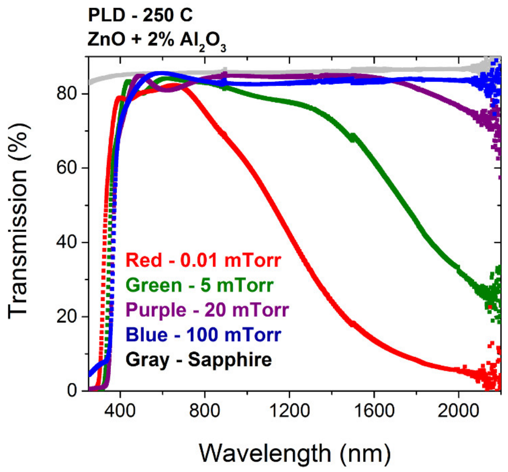
Utilization of Doped-ZnO and Related Materials Systems for Transparent Conducting Electrodes

M. Campion, H.L. Tuller in collaboration with A. Gougam, T. Buonassisi

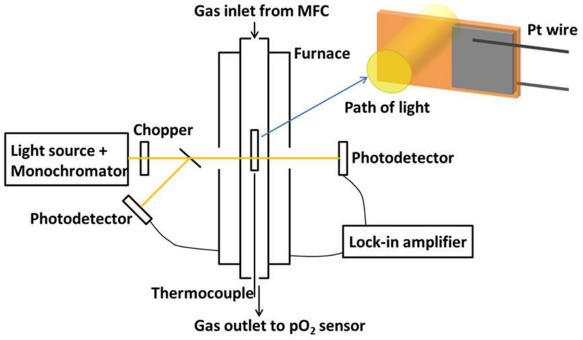
Sponsorship: Masdar Institute of Science and Technology

Efficient transparent electrode materials are vital for applications in smart window, LED display, and solar cell technologies. These materials must possess a wide band gap for minimal optical absorption in the visible spectrum while maintaining a high electrical conductivity. Tin-doped indium oxide (ITO) has been the industry standard for transparent electrodes, but limitations in both deposition temperature and use of the rare element indium has led to a search for better material alternatives. Doped ZnO represents one of the most promising alternatives, but the mechanisms by which processing conditions and defect chemistry affect the final material properties are not well understood. Reported values of the electrical and optical properties for doped ZnO can vary widely for seemingly similar processing conditions performed by different experimental groups. This could be due to the strong dependence on oxygen partial pressure, as demonstrated in Figure 1.

This work seeks to better understand the relationships between processing, defect chemistry, and material properties of ZnO. To accomplish these goals, methods such as *in situ* resistance and impedance monitoring during annealing and atom probe tomography will be applied. In addition, a variety of novel methods such as the *in situ* monitoring of optical transmission (shown in Figure 2) during annealing and the *in situ* monitoring of resistance during physical vapor deposition will be utilized to investigate ZnO. Direct measurements of the key constants for the thermodynamics and kinetics of oxidation in donor-doped ZnO will be experimentally determined for the first time. This increase in understanding will provide a predictive model for determining optical properties, carrier concentrations, and electron mobilities in ZnO, which is becoming an increasingly important material for transparent electrodes, nanostructures, and oxide transistors.



▲ Figure 1: Change in optical transmission characteristics of Al-doped ZnO due to changing oxygen partial pressure during deposition. This is brought about through the creation and annihilation of compensating ionic defects, which can heavily alter the carrier concentration.



▲ Figure 2: Schematic of experimental setup to be used for simultaneous *in situ* measurement of the optical transmission and electrical conductivity of thin film ZnO samples during annealing under controlled atmosphere and temperature.

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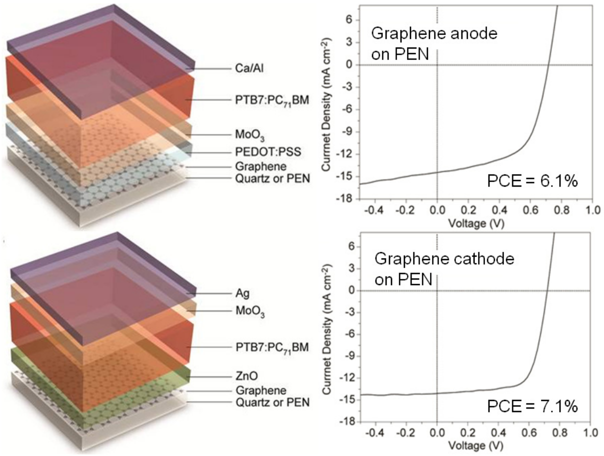
High-efficiency Graphene-based Flexible Organic Solar Cells

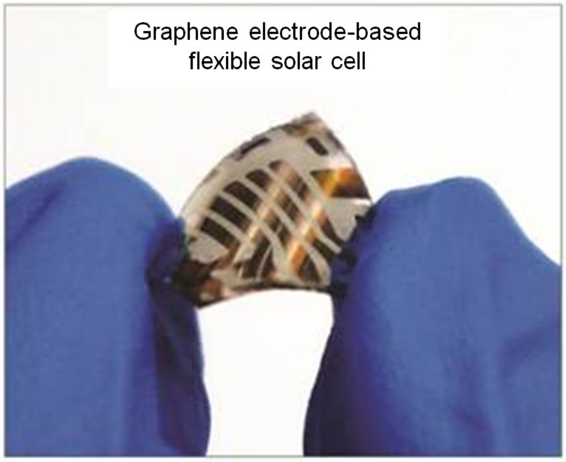
S. Chang, H. Park, X. Zhou, J. Kong, T. Palacios, S. Gradečak
Sponsorship: Eni S.p.A.

Flexible solar cells belong to the promising next generation of optoelectronic devices. Electrode materials with good conductivity, transparency, and flexibility must be developed for these solar cells. Graphene has been considered a promising flexible transparent electrode due to its good electrical conductivity and optical transparency along with mechanical and chemical robustness and potentially low-cost processing. We have successfully demonstrated both graphene anode- and cathode-based flexible polymer solar cells (PSCs) with thieno[3,4-b]thiophene/benzodithiophene (PTB7) and [6,6]-phenyl C71-butyric acid methyl ester ($PC_{71}BM$) by resolving the issues occurring at the interface between graphene and charge transporting semiconducting materials such as molybdenum trioxide (MoO_3) or ZnO. We demonstrate high-efficiency graphene electrode-based flexible PSCs with power conversion efficiencies of 6.1% (anode) and 7.1% (cathode). These efficiencies were achieved by thermal treatment of MoO_3 electron blocking layer and direct deposition of ZnO electron transporting layer on the graphene surface.

We expect our work to pave the way to realizing fully graphene electrode-based flexible solar cells by a simple and reproducible method. The advances accomplished in our work demonstrate graphene's promise as an alternative transparent electrode system in a variety of optoelectronic devices.



▲Figure 1: Schematic of a graphene anode- and cathode-based PSCs and corresponding AM1.5G *J–V* characteristics of graphene-based flexible PSCs.



▲Figure 2: A digital photograph of a flexible graphene PSC.

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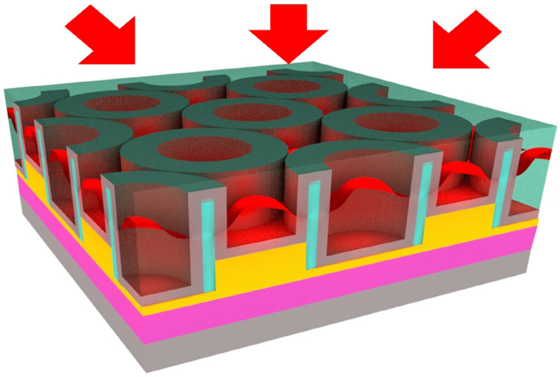
Enabling Ideal Solar-thermal Energy Conversion with Metallic Dielectric Photonic Crystals

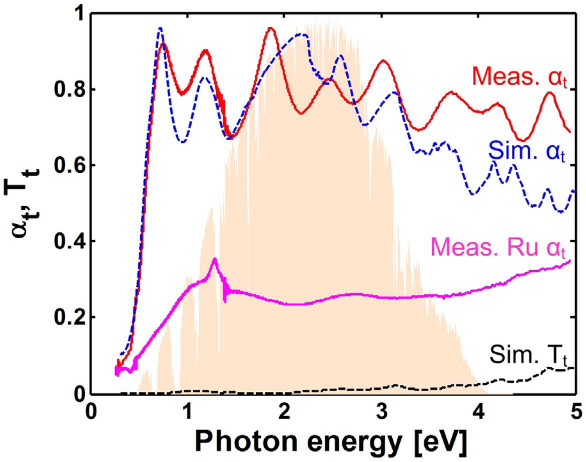
J.B. Chou, S.-G. Kim
Sponsorship: S3TEC

The selective absorption of sunlight plays a critical role in solar-thermophotovoltaic (STPV) energy conversion by tailoring both the absorption and emission spectra for efficient solar-thermal-electrical energy conversion. By selectively absorbing solar energy while suppressing long wavelength emission, optimal solar-thermal energy conversion can be achieved. In practical STPV systems, selective absorbers must simultaneously contain optical, manufacturing, and reliability properties. Here we present our solution, which contains all of the ideal properties of a selective absorber for large-scale and efficient solar energy conversion.

Metal absorption of sunlight plays a critical role in STPV energy conversion, selective solar absorption, selective thermal emission, and hot-electron generation. However, a broadband, high-temperature-stable, omnidirectional, wafer-scale, selective solar absorber from the visible to the near-IR has yet to be demonstrated experimentally. We present a silicon wafer-scale fabricated metallic dielectric photonic crystal (MDPhC) with an average absolute absorption of 85% for photon energies 5 eV>$\hbar\omega$>0.7 eV and an absorption below 10% for $\hbar\omega$<0.4 eV. Angled measurements show the existence of the cavity modes for angles up to 70° from normal. Furnace tests at 1000°C for 24 hours show no degradation in optical performance. Simulations indicate that the broadband absorption is due to a high density of the hybrid cavity and surface plasmon modes overlapped with an anti-reflection coating. The MDPhC was fabricated using traditional MEMS/CMOS-compatible fabrication techniques on a 6" wafer by utilizing the sidewall lithography technique. The optical properties of the MDPhC allow for broadband solar energy conversion and can lead to new applications for metal optics.



▲Figure 1: Schematic of the MDPhC device where incident light is coupled into cavity modes.



▲Figure 2: Measured absorption spectrum of MDPhC (red), simulated spectrum (blue), flat Ru (pink), and transmission through MDPhC (black). The solar spectrum is shown for reference.

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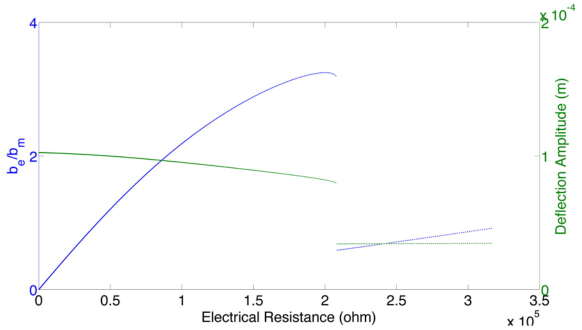
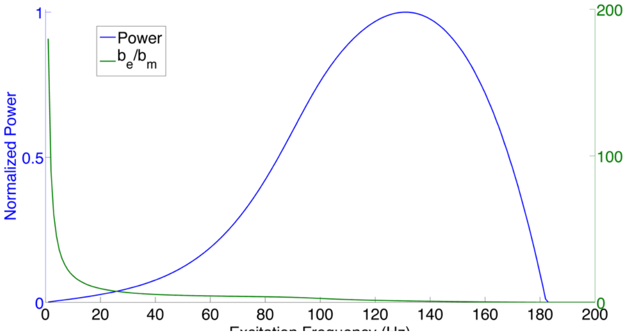
Nonlinear Resonance-based Piezoelectric Vibration Energy Harvesting

R. Xu, G. Gafforelli, Y. Yoon, A. Corigliano, S.-G. Kim

Linear resonance-based energy harvesters have been popular for vibration energy harvesting, due to their simplicity of micro-fabrication and high power efficiency at resonance. Nevertheless, the narrow frequency bandwidths that linear energy harvesters suffer prevent the technology from applying in the real frequency-changing ambient environment. As a promising potential solution, nonlinear resonance widens the power bandwidth by one order of magnitude, which shows the great potential of nonlinear designs. We built an electromechanically coupled, lumped model to provide a comprehensive analysis of nonlinear resonance-based energy harvesting. The model was based on the configuration of a doubly clamped beam with a thin film piezoelectric element working in d_{33} mode. The static indeterminate structure problem was solved with the Euler-Bernoulli beam theory and energy method. By considering the simple case of inputting a sinusoidal force and connecting the harvester to a resistor, we employed Kirchhoff's laws and the Harmonic Balance Method (HBM) to build and solve the nonlinear differential equations. Closed form expressions of the system's parameters were obtained from the analysis. The coupled, lumped model has verified varying electrical loads at each frequency to generate maximum power at that frequency by showing a power spectrum with a much wider bandwidth. Furthermore, the optimal electrical damping at each frequency was obtained; it shows that the electrical damping should be much higher than the mechanical damping to increase the power at low frequencies, and the maximum power is obtained when the electrical damping matches the mechanical damping.



▲Figure 1: The normalized electrical damping and deflection amplitude vs. electrical resistance. The insensitivity of the deflection to the change of damping explains the wide power bandwidth.



▲Figure 2: The maximum power envelope and the normalized optimal electrical damping at each frequency. The optimal electrical damping condition at each frequency could be useful to further widen the power bandwidth.

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Materials and Structures for Lithium-Air Batteries

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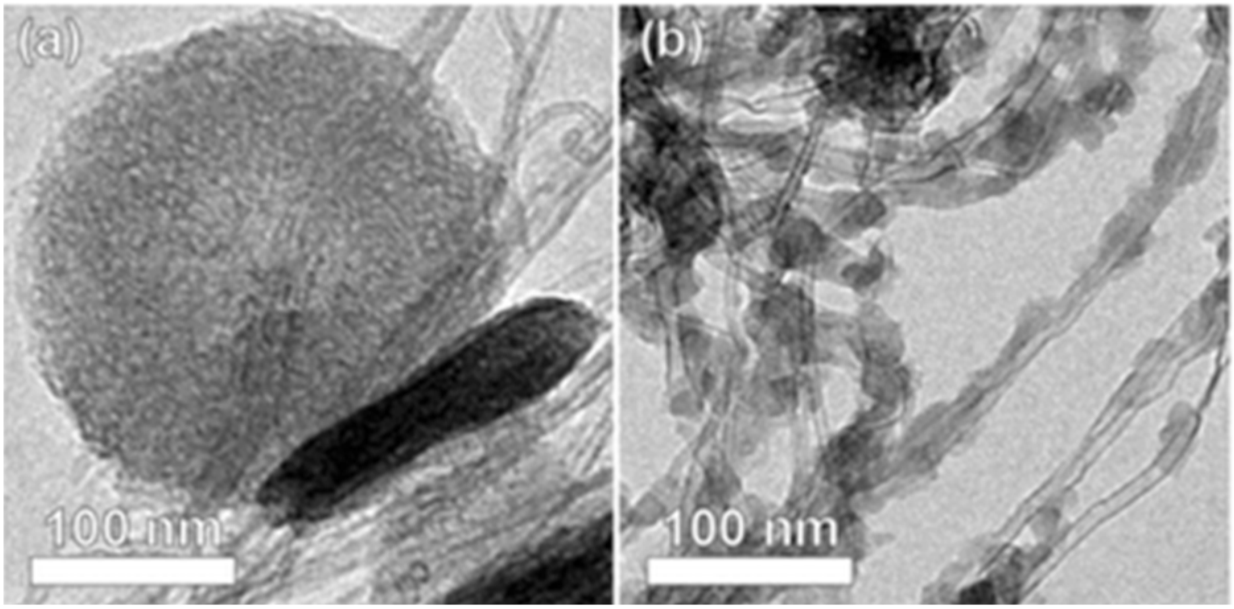
Sponsorship: Bosch, National Science Foundation, MRSEC

Lithium-air batteries hold promise for the next generation of electric vehicles and other applications. By reacting oxygen directly with lithium ions to form Li_2O_2 on discharge, these batteries can achieve energy densities 3-5 times higher than current lithium-ion batteries. However, a number of challenges exist for implementing lithium-air batteries, including poor rate capability, poor cyclability, high overpotentials upon charging, and electrode and electrolyte instability. We seek to address these issues by developing new electrode materials and architectures and performing studies of Li_2O_2 formation in different electrolyte solvents.

Aligned arrays of carbon nanotubes (CNTs) provide ideal conductive scaffolding materials for Li_2O_2, while occupying a small volume fraction and having low mass. CNTs of 5-10 nm in diameter are grown in aligned forests on catalyst deposited silicon wafer and delaminated. These free-standing carpets can be placed directly into our cell. We observed near-ideal gravimetric capacities and high volumetric capacities. We have observed two distinct types of morphologies upon discharge, depending on the rate and depth of discharge. At high rates of discharge, we observe the formation of copious small particles. At low rates and high depths of discharge, we observe the formation of large toroid particles (see Figure 1). From X-ray diffraction and selected area electron diffraction, we determined that these are Li_2O_2 particles with predominantly (0001) surfaces.

Additionally, we have studied the stability of these carbon nanotube electrodes in a lithium-air cell. We observed higher charging overpotentials as the cycle number increased. Using X-ray absorption near edge spectroscopy, we determined that Li_2CO_3 formed in increasing amounts upon cycling at the interface between lithium peroxide and the carbon nanotube. We attribute poor cycling performance and higher charge overpotentials to this Li_2CO_3 formation. We also observe the formation of LiOH in dimethyl sulfoxide solvent. We are currently working on comparative studies of different electrolyte and electrode materials to resolve these issues.



▲ Figure 1: TEM micrographs of Li2O2 formation at a) low rates and b) high rates.

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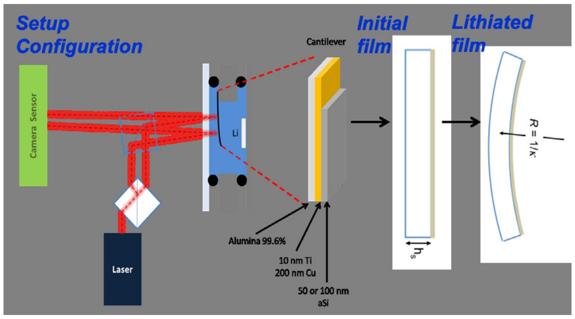
In situ Stress Measurements of Silicon Anodes in Lithium-ion Batteries

A. Al-Obeidi, S.T. Boles, R. Moenig, C.V. Thompson

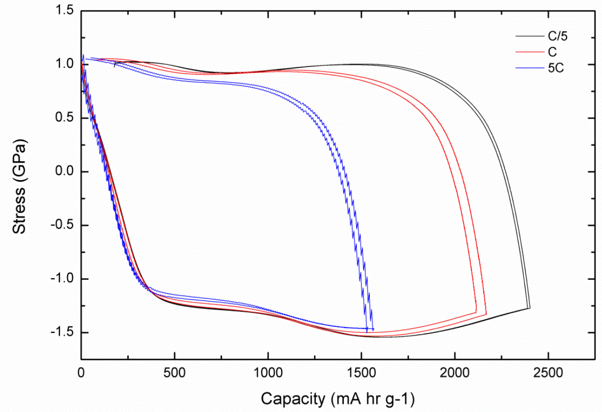
Sponsorship: SMART

There is increasing interest in developing autonomous microsystems, which integrate energy harvesting, energy storage, and power management in one package. Ideally suited for these applications are lithium-ion batteries with their relatively high energy-storage capacities. For the anode, we have investigated silicon, which offers among the highest known lithium capacity. However, accessing these large capacities results in large volume changes, rendering fully dense films or substrates unusable. To better understand the forces involved during cycling, *In situ* measurements of stress in silicon thin films (50 nm) were conducted using a home-built system.

During cycling, the silicon film expands during lithiation and contracts during delithiation. Using a cantilever setup, volume change in silicon results in cantilever bending, which can be quantified by laser deflection off the back of the cantilever (see Figure 1). *In situ* stress measurements reveal that during lithiation, there is a linear increase in stress (elastic regime) to ~ 1.2 GPa compressive. The stress then remains at about 1.2 GPa (plastic regime) to the end of lithiation. Upon delithiation, there is elastic unloading to 1 GPa tensile, and remains at that stress until the completion of delithiation. By monitoring the stress evolution of silicon electrodes during cycling allows one to probe the relationship between electrochemistry and mechanical behavior. This information provides new insight into designing nanostructured silicon films that minimize the stress during cycling.



▲Figure 1: *In situ* stress measurement setup to monitor the stress evolution of a 50-nm Si film during cycling.



▲Figure 2: Evolution of stress as a function of anode capacity at different cycling rates.

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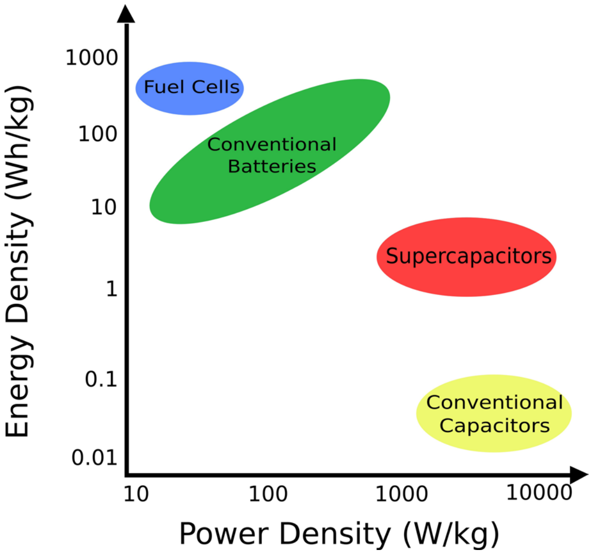
Graphene-based Supercapacitors for High-performance Energy Storage

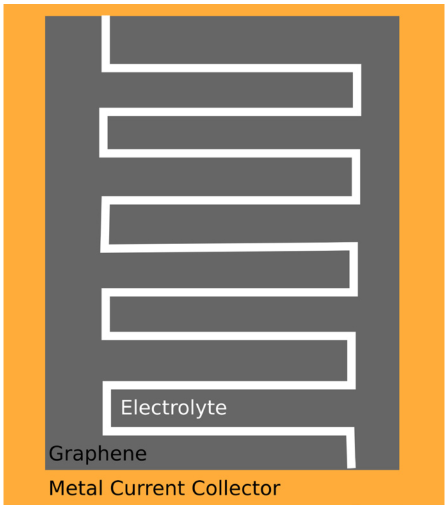
M. Aby, T. Palacios
Sponsorship: MIT-MTL Center for Graphene Devices and 2D Systems

As micro- and nanoscale devices get increasingly smaller and more advanced, supplying sufficient energy to them when needed is becoming more important than ever. Technology for energy storage has lagged behind other nanotechnologies, resulting in devices limited by the size and capabilities of their attached batteries. Electrochemical double-layer capacitors (EDLCs/supercapacitors) and relatively new carbon-based materials such as graphene offer the potential for large capacitances per unit volume, high specific power, high cycle life, rapid charge/discharge times, high efficiency, good performance in extreme conditions, and higher energy densities than conventional capacitors (see Figure 1). The supercapacitor structure allows for two layers of charge separation, one at each electrode, and its performance depends heavily on the choice of materials and on the amount of electrode surface area in contact with the electrolyte. Carbon-based materials, and graphene in particular, are especially promising for use as capacitor electrodes because of their high surface-area-to-volume and -weight ratios. Some work has already been done demonstrating the great promise of these graphene-based supercapacitors, but challenges still exist to scaling up their capacity and their production.

Our work aims to demonstrate supercapacitors based on graphene that are scaleable to energy-relevant dimensions for use in other devices and that can be produced relatively simply and inexpensively. We are currently exploring different ways of producing graphene-based supercapacitors by varying the type of graphene (chemical vapor deposition, reduced graphene-oxide, flakes, aerogels), the electrolyte material, the production method and the capacitor shape. Figure 2 shows an example of a typical supercapacitor structure, where the interlocked “fingers” of graphene electrodes are used to increase the surface area of graphene in contact with the electrolyte. This exploratory work will hopefully serve as a basis for future work in fabricating and scaling such devices and in integrating them with other technologies.



▲Figure 1: Energy density vs. power density comparison for energy storage devices. It is hoped that supercapacitors can eventually outperform current devices in both energy and power density.



▲Figure 2: Structure of an in-plane, graphene-based supercapacitor with solid-state gel-electrolyte. The electrolyte used is (PVA)-H_3PO_4.

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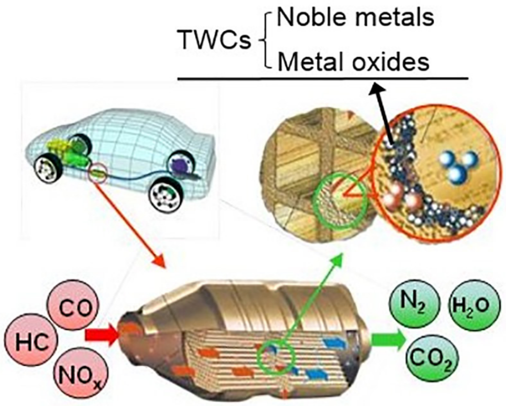
Catalytic Oxygen Storage Materials

Y. Cao, D. Chen, H. Saltzburg, S.R. Bishop, H.L. Tuller

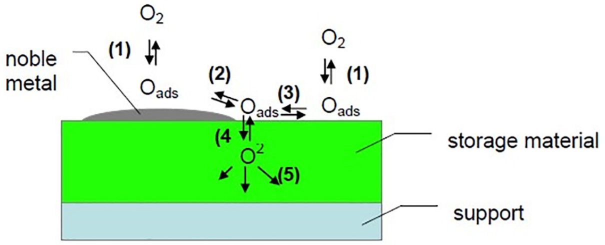
Sponsorship: Department of Energy, Basic Energy Science

The combustion of fossil fuels, essential for electricity generation and vehicular propulsion, is generally incomplete, leading to harmful NO_x, CO, and unburned hydrocarbon emissions. Progress in minimizing such emissions has relied on the operation of "three way catalysts" (TWCs, see Figure 1) which utilize a combination of noble metals and metal oxides with the ability to take up or release oxygen for reduction/oxidation of pollutants (NO_x to N_2 and CO & HC to CO_2 and H_2O, respectively). In this project, we investigate the rate at which oxygen storage materials (OSM, typically $Ce_xZr_{1-x}O_{2-\delta}$ and $Pr_xCe_{1-x}O_{2-\delta}$) exchange oxygen with the atmosphere and the magnitude of oxygen that they store with the aid of geometrically well-defined thin film and bulk structures (see Figure 2). We use impedance spectroscopy, Kelvin probe, thermogravimetry, Raman scattering, and electrical conductivity measurement methods to determine electrochemical performance and oxygen storage capabilities. Based on the measurements of oxygen non-stoichiometry and electrical conductivity, we have developed defect equilibrium models.

Possible interactions between the noble metals and the metal oxide components in TWCs influence the catalytic activity. The role of the state of OSM and its interactions with catalytic metals will be studied by analyzing *in situ* electrical conductivity simultaneously with catalytic reactions. The electrical properties, when correlated to actual TWC performance, using a differential flow reactor, will allow for a more detailed understanding of performance criteria.



▲Figure 1: Application of TWCs in the automobile.



▲Figure 2: Microscopic processes involved in oxygen storage: oxygen ad- and desorption (1), spillover and backspillover (2), surface diffusion between surface exchange and adsorption sites (3), oxygen incorporation and release (4), and bulk diffusion (5).

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Influence of Strain on Ionic Conduction

S.N. Cook, H.L. Tuller

Sponsors: Department of Energy, Basic Energy Science

Ionic conduction in ceramic materials plays a vital role in a wide range of devices in fields as diverse as clean energy generation and healthcare. Device implementation is often hindered by the low inherent conduction levels present at low temperatures, even in highly chemically optimized materials, a side effect of having to move large ions through a relatively static solid state material. Recent reports suggest that ionic conduction properties can be significantly enhanced through the fabrication of nanostructures. The interpretation of these results remains controversial, but it has been suggested that strain due to lattice mismatch at heterointerfaces has the greatest effect. This phenomenon has never been directly measured due to the great deal of complexity inherent in thin film nanostructures.

In this project we take an alternative approach to the isolation of this effect through the direct mechanical manipulation of the strain state of fast ion conducting ceramics using a novel experimental setup. Developing a full understanding of the relationship between strain and ionic conduction will not only allow for devices that take advantage of this relationship, one potential application being high temperature strain gauges, but also allow developments in current device technology where material optimization takes into account chemistry and microstructure, as well as the strain state, facilitating superior device performance and increasing operating condition range.

Development of Reversible Solid Oxide Cells: A Search for New Electrode Materials

C.S. Kim, H.L. Tuller

Sponsorship: Skolkovo Foundation

Recent technological advances in photovoltaic cells have enabled efficient conversion of solar energy to electricity at low cost. However, solar radiation varies throughout the day, and excess electricity generated during the daytime is lost if not used. Electrolysis cells can convert the excess electrical energy into chemical fuels via electrolyzing water (H_2O) or carbon dioxide (CO_2) to hydrogen (H_2) or carbon monoxide (CO), respectively. The excess energy stored in chemical form can be converted back to electricity by operating solid oxide fuel cells (SOFCs). Conversion of energy into chemical fuels via electrolysis is advantageous over storage in batteries given potentially reduced costs and the high energy density of chemical fuels. A reversible SOFC can operate as an electrolysis cell during the day, and as a fuel cell at night. It has been shown that typical SOFCs can be efficiently operated as both fuel cells and electrolysis cells, but with significant degradation. This degradation is mainly due to the redox instability of the electrodes in reverse operating mode. While symmetric solid oxide cells with redox stable electrodes have been investigated, their performance was substantially lower than the state-of-the-art SOFCs. In this project, we are investigating new redox stable electrode materials with high mixed (electrical and ionic) conductivity in both highly oxidizing and reducing conditions. Half-cells and symmetric cells are fabricated by depositing the new electrode material onto an electrolyte by pulsed laser deposition. Structural analysis of the new material is conducted by high-resolution X-ray diffraction and electro-chemical properties are measured by means of impedance spectroscopy in differing temperatures and atmospheres (pO_2 and pH_2).

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Investigation of Fuel Cell Cathode Performance in Solid Oxide Fuel Cells: Application of Model Thin Film Structures

J.J. Kim, D. Chen, N. Perry, N. Thompson, S.R. Bishop, H.L. Tuller

Sponsorship: Department of Energy

Understanding the reaction kinetics controlling the oxygen reduction reaction (ORR) between the gas phase and the oxide lattice in solid oxide fuel cell (SOFC) cathodes is considered to be critical for achieving improved device performance, especially at reduced operating temperatures. Although numerous research activities have been focused on elucidating the electrode reaction mechanisms, their conclusions remain unsatisfactory and controversial. The ORR at mixed conducting oxide thin film cathodes consists of oxygen adsorption, dissociation, charge-transfer, and incorporation steps. The kinetic parameters associated with the overall ORR, such as the diffusion coefficient (*D*) and surface exchange coefficient (*k*), are strongly influenced by the oxygen nonstoichiometry, δ, of the oxide. Because many advanced oxide materials used in SOFC experience significant changes in δ during operation at elevated temperatures and under reducing/oxidizing conditions, the ability to diagnose a material's behavior in operando is therefore important.

Our group recently demonstrated that δ in $Pr_{0.1}Ce_{0.9}O_{2-\delta}$ (10 PCO) thin films could be reliably derived by utilizing chemical capacitance extracted from electrochemical impedance spectroscopy (EIS) measurements. Furthermore, we introduced a non-contact optical means for *in situ* recording of transient redox kinetics, as well as the equilibrium Pr oxidation state and, in turn, δ in 10 PCO thin films, by monitoring the change in absorption spectra upon change in pO_2 or temperature. In this study, we are investigating cathode kinetics and nonstoichiometry of two model oxide thin films; $Ba_xSr_{1-x}Ti_{1-y}Fe_yO_{3-y/2+\delta}$ (BSTF) and $Pr_xCe_{1-x}O_{2-\delta}$ (PCO) by simultaneously utilizing *in situ* optical absorption spectroscopy and EIS as a function of temperature, pO_2 and electrical potential. We are also investigating changes in surface chemistry and their impact on electrode impedance by atomic force microscopy (AFM), X-ray photoelectron spectroscopy (XPS), and low-energy ion scattering spectroscopy (LEIS).

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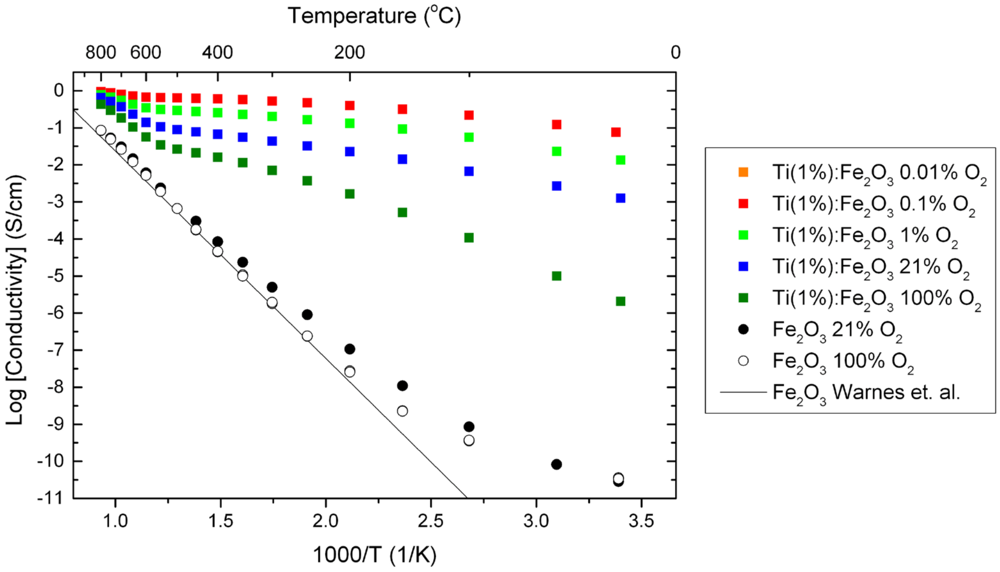
Direct Solar-to-hydrogen Conversion: Low-cost Photoelectrodes

J. Engel, H.L. Tuller, in collaboration with R. van de Krol

Sponsorship: MIT Energy Initiative

With continuously growing energy demands, alternative emission-free solar energy solutions become ever more attractive. However, to achieve sustainability, efficient conversion and storage of solar energy is imperative. Photoelectrolysis harnesses solar energy to evolve hydrogen and oxygen from water, thereby enabling energy storage via chemical means. In this work, we investigate photoelectrodes that offer high conversion efficiency, long-term stability, and low cost,. The focus is on semiconducting metal oxides, in which the energy bands, the defect, and microstructures are tuned to optimize optical absorption, charge transport, and reduce overpotentials. For high efficiency, transition metal-based oxidation catalysts are implemented at the photoelectrode.

Hematite, or α-Fe_2O_3, has emerged as a highly promising photoanode candidate for photoelectrochemical cells. While significant improvements in its performance have recently been achieved, the reason that the maximum photocurrents remain well below their theoretical predictions remains unclear. We report a detailed correlation between the electrical conductivity of undoped and 1 atom% Ti doped hematite and their annealing conditions (varying temperature and partial pressure of oxygen). Hematite thin films grown by pulsed laser deposition onto sapphire single crystals are evaluated by impedance spectroscopy. Hematite's room temperature conductivity can be increased from $\sim 10^{-11}$ S/cm for undoped films by as much as nine orders of magnitude by doping with the Ti donor. Furthermore, by controlling the non-stoichiometry of Ti-doped hematite, one can tune its conductivity by up to five orders of magnitude. Depending on processing conditions, donor dopants in hematite may be compensated for largely by electrons or by ionic defects (Fe vacancies). We derive a defect model to explain this phenomenon. These results highlight the importance of carefully controlling photoanode processing conditions, even when operating within the material's extrinsic dopant regime, and more generally, provide a model for the electronic properties of semiconducting metal oxide photoanodes.



▲ Figure 1: Conductivity of Ti doped and undoped α-Fe_2O_3 as a function of temperature at different pO_2s.

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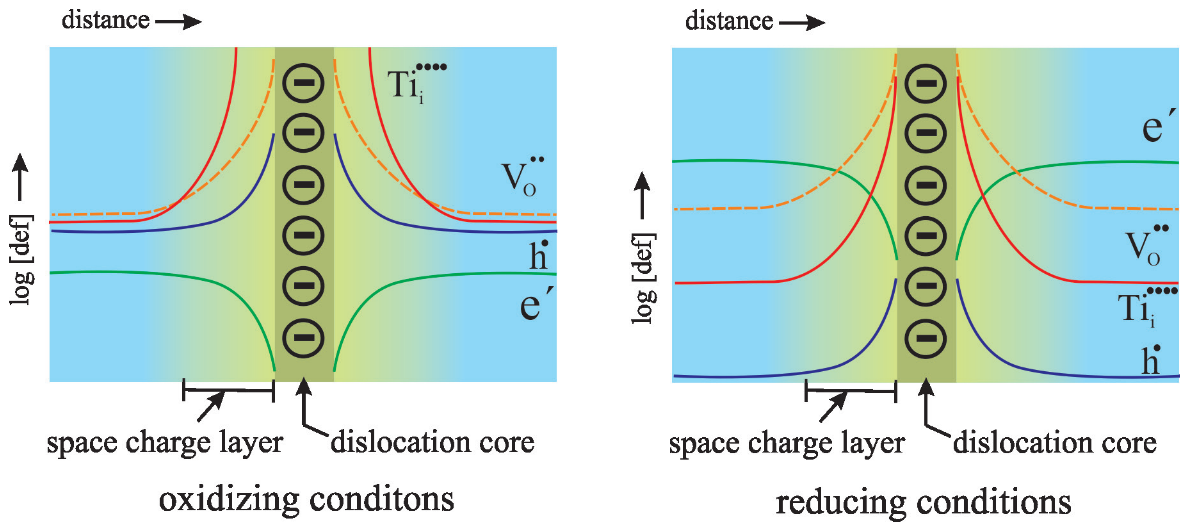
Influence of Mechanical and Electrical Effects on Ionic and Electronic Defect Transport

K.K. Adepalli, B. Yildiz, H.L. Tuller

Sponsorship: Basic Energy Sciences, Department of Energy; MIT Center for Materials Science and Engineering, National Science Foundation

Defects are essential for charge and mass transport in ionic solids. Altering the defect concentration and/or the charge states therefore leads to modification of the transport properties in ionic solids. Hence, aliovalent doping and morphology (grain boundary and/or nano-size effects) engineering have been the topic of interest for several decades. In addition to these zero-dimensional point defects (dopants) and two-dimensional morphological defects (grain boundaries), one-dimensional line defects (or dislocations) can also potentially influence transport properties. Interestingly, a fundamental understanding of the effect of these defects on charge transport is still lacking in ionic solids. A key feature of dislocations in ionic solids is that they are often charged due to a lower defect formation energy and a higher defect segregation driving force in comparison to the bulk. Dislocations also result in a variation in local strain state (compressive or tensile) that can influence defect mobility. Due to excess charge on the dislocation core and the adjacent defect redistribution (space charge zones, see Figure 1) and the locally strained lattice, these defect sites are believed to be electrochemically active centers for catalysis and resistive switching memory behavior through the preferential formation of highly conductive filaments along dislocations. With the understanding of point defect transport as the common thread, this project focuses on these two emerging fields. The project incorporates the investigation of dislocation effects (by mechanical means) on catalytic properties of solid oxide fuel cell cathode materials. In addition, electric field effects on point defect transport will be investigated, not only to find new memristor devices based on electrochemical switching, but also to investigate the underlying mechanism of charge transfer under an applied electric field.



▲Figure 1: Schematic representation of dislocation core charge and adjacent defect redistribution in space charge regions in TiO_2 under oxidizing and reducing conditions.

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Ionic Conduction Studies in TlBr Radiation Detector Materials

S.N. Cook, S.R. Bishop, H.L. Tuller in collaboration with S. Payne, A. Conway, K. Shah

Sponsors: Domestic Nuclear Detection Office, Lawrence Livermore National Laboratory, Radiation Monitoring Devices, Inc.

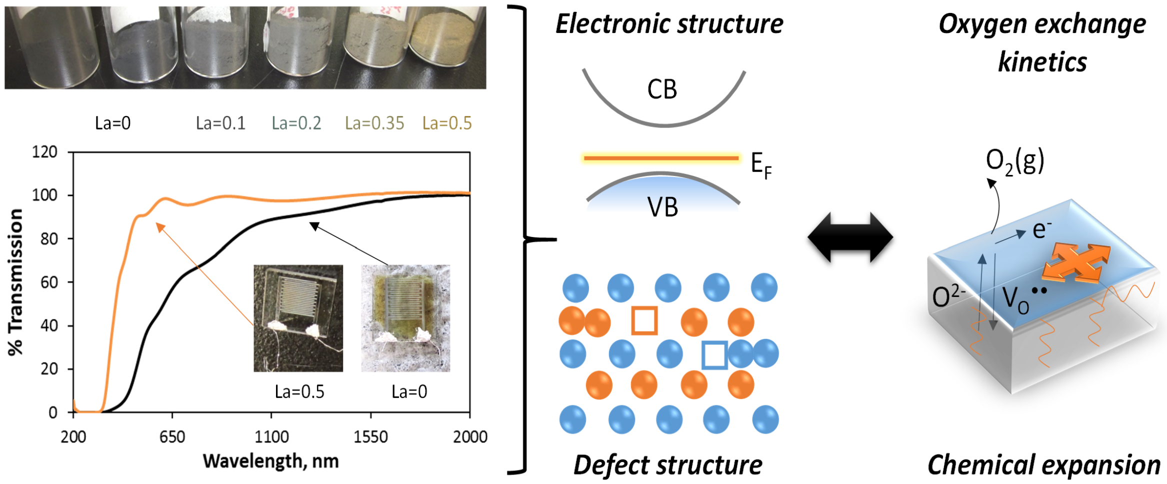
Detection of high-energy radiation (e.g., γ-rays) is key in nuclear non-proliferation strategies. When a wide-band gap semiconductor detector intercepts a γ-ray, electron-hole pairs are formed, resulting in an increase in electrical conductivity, facilitating their detection. As in any photodetecting device, sensitivity is maximized if the conductivity in the non-illuminated (dark) state is very low. While current semiconductor-based technologies require cooling to very low temperatures, adding to cost and reducing portability, TlBr is an attractive detector material given its low room temperature dark conductivity, as well as its high mass density, leading to high radiation absorption. In TlBr the dominant conduction mechanisms, when non-illuminated, are ionic in nature. Their minimization is therefore valuable to produce higher performance detectors. In this project, we characterize the ionic conduction properties of TlBr, dopant association and exsolution using impedance spectroscopy. Through doping techniques, we have determined that TlBr is primarily a Schottky type ionic conductor, meaning that Tl and Br move through the material by vacancy motion. These measurements have led us to predict a doping strategy to minimize dark conductivity. Further limiting device adoption are short operating times of less than 100 days. We are currently implementing the detailed defect model that we have established by investigating the origin of this long-term performance degradation and its relation to electrode chemistry.

Fundamental Studies of Oxygen Exchange and Associated Expansion in Solid Oxide Fuel Cell Cathodes

N.H. Perry, J.J. Kim, D. Marrocchelli, J.E. Thomas, S.R. Bishop, H.L. Tuller in collaboration with B. Yildiz, D. Pergolesi
Sponsorship: Department of Energy

To lower the cost of solid oxide fuel cells (SOFCs), both the low-temperature efficiency and long-term durability need to be improved. Fundamental studies of how fuel cell materials "breathe" oxygen under operating conditions can address these dual needs, since: 1) sluggish oxygen incorporation at the cathode dominates SOFC efficiency losses at low temperatures and 2) chemical expansion during oxygen loss from the oxide can result in catastrophic mechanical failure of the cell. Our recent review article highlights the widespread presence of chemical expansion and its consequences across a number of energy conversion and storage devices.

In this work we experimentally and theoretically investigate cathode systems with model geometries (controlled active surface area and diffusion lengths of thin films) and model chemistries (tailored electronic structure, crystal structure, and defect chemistry) to isolate underlying factors controlling the oxygen exchange and expansion behavior. Previous work on fluorite-structured electrodes is being extended to the perovskite families (Sr,Ba,Ca,La)(Ti,Fe,Co)$O_{3-\delta}$, and (La,Sr)(Ga,Ni)$O_{3-\delta}$, using advanced *in situ* X-ray diffraction, optical absorption, thermogravimetric analysis, dilatometry, electrochemical impedance spectroscopy techniques, defect thermodynamic modeling, and density functional theory calculations. Using this approach we have experimentally confirmed our previous theoretical calculations demonstrating the important role of charge localization in controlling chemical expansion behavior. We have also identified how chemical substitution affects defect chemistry, electronic structure, and corresponding oxygen exchange rate at the cathode surface. This performance over time is being correlated to changes in surface chemistry, studied by high-resolution energy dispersive spectroscopy of thin film cathodes and low-energy ion scattering. Such information is key to the design of both efficient and durable fuel cell electrodes.



▲ Figure 1: Optical properties (left) serve as markers for electronic and defect structures, to determine their relationship to the oxygen exchange kinetics and corresponding expansion of solid oxide fuel cell cathodes.

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A Universal Village: Our Desired Living Conditions

Y. Fang, B.K.P. Horn, I. Masaki
Sponsorship: Intelligent Transportation Research Center, MTL

The worldwide trend of urbanization makes more people live a "modern lifestyle" that revolves around vehicles and consumption of natural resources faster than the rest of the world, which intensifies the environmental and sustainability crisis. The current society is facing serious challenges due to fast urbanization, limited resources, and the increasing population of senior citizens. While there have been efforts to address challenges to humanity up now, many current designs are availability-based, technique-oriented, and bottom-up schemes that focus on individual elements. The whole living environment lacks a coordinated and systematic design and runs into new challenges including new security, safety, and privacy concerns plus environmental and sustainability problems. Right now, transportation is the second cause, after electricity generation, of the CO2 problem. Instead of passively responding to different emerging problems with temporary solutions, people in different fields should work together to find systematic long-term solutions for the future.

The Universal Village (UV) is a new concept proposed by MIT Universal Village Program, an expanded and advanced version of Smart Cities that pursues human-nature harmony through the wise use of technologies. The concept is named for two considerations. First, "smart" is not the final objective. Instead, the desired future society should follow laws of the universe and protect the environment and ecosystem. Otherwise, Mother Nature suffers and we humans suffer eventually. Second, cities might not be the only living environment in our future. Universal Villages are multi-functional, multi-format, and sustainable communities that incorporate the ideal mixture of cities and suburban areas.

The development of the UV follows a need-based, human-nature-oriented, and systematic top-down design approach. The UV calls for a collective effort from multidisciplinary fields and new innovative technologies that would change our future lives and improve human-nature harmony in the long run. The UV is expected to provide sustainable happiness to residents, so we must understand humans' need in depth before developing intelligent technologies to improve comfort, convenience, and efficiency for human beings. We expect to develop both sensing technologies to understand humans' behavior and cities' activities and integrated system solutions to provide intelligent reactions at various levels in cities. A better understanding of family routines and human activities in buildings will benefit the development of intelligent safety monitoring and intelligent energy saving. Real-time detection of traffic situations will allow control centers to respond to accidents faster than before. Intelligent health monitoring systems, distributed medical treatment, and tele-healthcare can provide early warnings before people physically feel sick and diseases spread widely. We aim to propose new lifestyles that are in harmony with nature and need collective efforts from multi-disciplinary research. The major themes include the following:

- Innovative technologies that support new lifestyles for future transportation, city management, healthcare, etc.
- Integrated solutions to urgent and long-term challenges of safety, security, privacy, climate changes, etc.
- Environmentally friendly energy sources such as solar power, bio-energy, supergrids, wireless charging, etc., and
- Environmentally friendly new materials, e.g., pesticides/fertilizers/etc. without chemical contamination, and decomposable utensils, tools, etc. without extra burdens for garbage disposal.

FURTHER READING

- Y. Fang, B. K. P. Horn, and I. Masaki, "Universal Village: What? Why? How?" presented at *2013 International Conference on Universal Village.*
- Y. Fang, S. Yokomitsu, B. Horn, and I. Masaki, "A Layered-based Fusion-based Approach to Detect and Track the Movements of Pedestrians through Partially Occluded Situations," presented at *IEEE Intelligent Vehicles Symposium 2009* (IV2009).
- B. Horn, Y. Fang, and I. Masaki, "Hierarchical framework for direct gradient-based time-to-contact estimation," presented at *IEEE Intelligent Vehicles Symposium 2009* (IV2009).