

Thin Films Division

Room A122-123 - Session TF+EM+NS+SS-ThM

Thin Films for Energy Harvesting and Conversion

Moderators: Siamak Nejati, University of Nebraska-Lincoln, Xinwei Wang, Shenzhen Graduate School, Peking University

8:00am TF+EM+NS+SS-ThM1 Redesigning Batteries into Efficient Energy Harvesters and Sensors for Wearable Applications, *Cary Pint*, Vanderbilt University **INVITED**

Here I will discuss the research efforts of my team demonstrating how active materials utilized in batteries can be reconfigured into an electrochemical framework to harvest, rather than store, energy. This new functionality of battery materials arises from the fundamental coupling between mechanical stresses and electrochemistry that my group has demonstrated while investigating the "strain-engineering" of battery materials. By exploiting this coupling in a symmetric cell device configuration, we are able to construct devices that convert mechanical energy to electrical energy by mechanical modulation of the electrochemical reaction potential. I will discuss the development of this device platform from proof-of-concept device fabrication using 2D materials to our most recent demonstration of textile-integrated biocompatible fibers integrated into fabrics for harvesting/sensing of human motion. Most notably, I will discuss how the sluggish diffusion kinetics of ions between two electrodes – whereas a challenge for emerging battery applications, enables these devices to measure a continuous response from the whole broad range of frequencies associated with human motion. This allows these wearable harvesters to provide real-time sensing data that can be directly correlated with dynamic human motion models. This new approach leverages the efficient nature of electrochemistry, the wide range of materials selection and chemistries relevant for batteries, and without any of the safety concerns of batteries due to the symmetric electrode configuration.

8:40am TF+EM+NS+SS-ThM3 Engineering Effective Back Contact Barrier by interfacial MoSe₂ defect states for CZTSe: nanolayer Ge solar cells, *Sanghyun Lee*, Indiana State University

The steadily emerging Cu₂ZnSnS₄ (CZTSSe) devices are alternative thin film solar cells with abundant elements in earth's crust for the past several years. Despite several advantages such as high absorption coefficient (>10⁴ cm⁻¹) and a tunable direct band gap energy (1 to 1.4 eV), the improvement and understanding have been stagnant in the past several years. Recently, CZTSe: nanolayer Ge solar cells have shown significantly improved pseudo-mono grain toward the depth direction.

Due to the improvement and the similarity between CZTSe and Cu(In,Ga)Se₂ (CIGS) thin film solar cells, the CZTSe/Molybdenum (Mo) back contact interface was often misinterpreted by expecting the similar back contact property to CIGS. However, unlike the stable CIGS (CuInSe₂)/Mo interface, the CZTSe/Mo interface is thermodynamically unstable due to the higher oxidation states of Sn. Although the presence of an interfacial MoSe₂ layer at Mo/absorber is always confirmed, properties of the back contact-interface such as structure and electrical behaviors are convoluted.

Following our empirical results about the back contact barrier of CZTSe: nanolayer Ge devices, we perform analytical and numerical modeling to explain the back contact improvement theoretically. The device modeling are carried out with the simulator, developed at Indiana State University. The tool is run in MATLAB environment, connected to other external tools (Sentaurus TCAD). Based on our result, defects in MoSe₂ interfacial layer dominate the back contact property of CZTSe: nanolayer Ge devices by increasing of the effective back contact barrier, which consists of two different back contact barriers, thereby increasing series resistance as well. The reduction of MoSe₂ defect concentration from 1 × 10¹⁷ to 1 × 10¹⁵ cm⁻³ decreases the effective barrier height by 51 meV, which results in approximately 34 % decrease in the series resistance (See supporting data). Conversely, as the defect concentration increases, the benefit from the back contact barrier lowering by the valence bands offset between MoSe₂ and CZTSe absorber is reduced and essentially eliminated. However, the back contact barrier between MoSe₂ and Mo metal contact remains the same even with increased MoSe₂ defect concentration. Incorporating thin Ge nanolayer at the interface between the absorber and MoSe₂ positively influences and possibly reduces the defect states, lowering the effective barrier. The exponential fitting of the effective barrier and series resistance agrees well with the experimental results. The improvement of the back

contact barrier for CZTSe: nanolayer Ge devices is calculated as 23.8 meV than CZTSe without nanolayer Ge devices.

9:00am TF+EM+NS+SS-ThM4 Development of Low-Cost, Crack-Tolerant Metallization Using Screen Printing for Increased Durability of Silicon Solar Cell Modules, *O.K. Abudayyeh*, *Osazda Energy*; *A. Chavez*, University of New Mexico; *J. Chavez*, *Osazda Energy*; *Sang M. Han*, University of New Mexico; *F. Zimbardi*, *B. Rounsaville*, *V. Upadhyaya*, *A. Rohatgi*, Georgia Institute of Technology; *B. McDanold*, *T. Silverman*, National Renewable Energy Laboratory

One of the ways to reduce the cost of solar electricity to 3¢/kWh, thus reaching parity with fossil-fuel-based generation, is to reduce the degradation rate of solar modules and extend their lifetime well beyond 30 years. The extended module lifetime in turn can positively influence the financial model and the bankability of utility-scale PV projects. Today, the highest-risk-priority solar module degradation mechanism is what is known as hot spots, often induced by cell cracks. In order to address this degradation mechanism, we make use of low-cost, multi-walled carbon nanotubes embedded in commercial screen-printable silver pastes, also known as metal matrix composites. When the carbon nanotubes are properly functionalized and appropriately incorporated into commercial silver pastes, the resulting metal contacts on solar cells, after screen-printing and firing, show exceptional fracture toughness. These composite metal contacts possess increased ductility, electrical gap-bridging capability up to 50 μm, and "self-healing" to regain electrical continuity even after cycles of complete electrical failure under extreme strain [1]. Accelerated thermal cycling tests on mini-modules constructed from aluminum back surface field (Al-BSF) cells show a slower degradation rate for the cells integrated with the composite grid fingers and busbars for the front surface metallization compared to the cells with conventional metallization.

[1] O. K. Abudayyeh, A. Chavez, J. Chavez, S. M. Han, F. Zimbardi, B. Rounsaville, V. Upadhyaya, A. Rohatgi, B. McDanold, T. J. Silverman, and N. Bosco, in "Low-Cost Advanced Metallization to Reduce Cell-Crack-Induced Degradation for Increased Module Reliability," 2019 NREL PV Reliability Workshop, Lakewood, CO, 2019.

9:20am TF+EM+NS+SS-ThM5 Fabrication of Optical Test Structures for Enhanced Absorption in Thin Multi-junction Solar Cells, *Erin Cleveland*, *N.A. Kotulak*, *S. Tomasulo*, *P. Jenkins*, U.S. Naval Research Laboratory; *A. Mellor*, *P. Pearce*, Imperial College London, UK; *N.J. Ekins-Daukes*, University of New South Wales, Australia; *M.K. Yakes*, U.S. Naval Research Laboratory

In space applications, a key figure of merit is conversion efficiency at end-of-life, which combines both beginning-of-life efficiency with degradation due to radiation exposure on orbit. In currently used InGaP/GaAs/Ge triple junctions, the GaAs middle cell has the most pronounced degradation, which limits the total current generation at the end-of-life. Recently, we demonstrated that as the thickness of the GaAs cell decreases, the tolerance to radiation damage increases. [1] However, because the cell absorbs less light as the thickness of the active region is reduced, the beginning-of-life performance suffers as compared to optically thick cells. To realize the benefits of both structures, light trapping architectures may be used to increase absorption within the cell while still maintaining the increased radiation tolerance of the thinner geometry.

Designing a wavelength selective light trapping structure positioned interstitially between two of the subcells of a multi-junction device is a new challenge which prohibits many of the well-known light trapping techniques. Recently, we have proposed a structure which combines a distributed Bragg reflector (DBR) with a textured diffraction grating. [2] Such a structure would provide substantial absorption of light in the middle subcell of a multi-junction device, while still allowing enough low-energy light to pass through the structure so the bottom cell remains well current matched with the other junctions. This structure is proposed to have over an order of magnitude increase in overall radiation tolerance while maintaining comparable beginning of life performance to the current technology.

In this presentation, we present a first experimental demonstration of this structure. The design combines a diffraction grating fabricated via nanosphere lithography [3], a low-index transparent spacer layer, and a DBR, which synergistically traps light inside the targeted subcell. This presentation will highlight processing techniques and challenges associated with fabricating a textured ultra-thin solar cell, while illustrating the effectiveness of integrating light trapping structures within an ultra-thin

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solar cell as an effort towards realizing high efficiency ultra-thin photovoltaic devices.

[1] L. C. Hirst, *et al.*, "Intrinsic radiation tolerance of ultra-thin GaAs solar cells", *APL*, 109 (2016)

[2] A. Mellor, N.P. Hylton, S.A. Maier, N. Ekins-Daukes, "Interstitial light-trapping design for multi-junction solar cells", *Solar Energy Materials & Solar Cells*, 159, (2017)

[3] H.W. Deckman and J.H. Dunsmuir, "Natural lithography", *Applied Physics Letters*, 41(4) (1982)

9:40am **TF+EM+NS+SS-ThM6 Phosphorus as a *p*-Dopant in Pyrite FeS₂, a Potential Low-cost earth-abundant Thin Film Solar Absorber**, *Bryan Voigt*¹, W. Moore, D. Ray, M. Manna, University of Minnesota, Minneapolis; J.D. Jeremiason, Gustavus Adolphus College; L. Gagliardi, E.S. Aydil, C. Leighton, University of Minnesota, Minneapolis

Pyrite FeS₂ has long been considered an ideal absorber material for low-cost and sustainable thin film solar cells because it is composed of earth-abundant, non-toxic, inexpensive elements, has a suitable band gap (0.95 eV), and absorbs light so strongly that a 100-nm-thick film absorbs >90 % of photons with energies above the band gap. Lack of doping control, however, has presented a barrier to realization of the *p-n* pyrite homojunction, *i.e.*, the simplest route to a pyrite solar cell. *Heterojunction* pyrite solar cells have proven to have disappointingly low efficiencies (~3%), surface conduction and leaky surface inversion layers being implicated as the culprit. While mitigation of pyrite surface conduction remains a challenge, doping has begun to yield to understanding, renewing optimism for a *p-n* pyrite homojunction solar cell. In particular, we have shown that rigorously phase-pure pyrite single crystals and thin films are exclusively *n*-type, due to a common dopant. Most recently, we have identified sulfur vacancies as this unintentional *n*-dopant, enabling robust control over *n*-doping levels in single crystals grown by chemical vapor transport (CVT). Progressing towards a *p-n* pyrite homojunction, here we demonstrate effective *p*-type doping in crystals by introducing phosphorus in the vapor phase during CVT growth. Increasing the phosphorus concentration from <0.1 ppm to 30 ppm evolves electronic conduction from *n*-type to *p*-type, with a clear and reproducible majority carrier inversion for concentrations >10 ppm. Typical transport properties of phosphorus-doped, *p*-type pyrite crystals include a hole thermal activation energy, room temperature resistivity, hole density, and mobility of ~170 meV, 3 Ω cm, 2×10^{18} cm⁻³, and 1 cm² V⁻¹s⁻¹, respectively. Density functional theory calculations confirm that phosphorus substituted on the S site is an acceptor, predicting a defect level at 200 meV above the valence band maximum, in good agreement with experiment. With both *n*- and *p*-type doping control achieved, attempts at *p-n* pyrite homojunction solar cells become possible.

This work was supported by the customers of Xcel Energy through a grant from the Renewables Development Fund and in part by the National Science Foundation through the University of Minnesota MRSEC under DMR-1420013.

11:00am **TF+EM+NS+SS-ThM10 Relaxor-ferroelectric Thin Films for Energy Harvesting from Low-grade Waste-heat**, *Amrit Sharma*, B. Xiao, S.K. Pradhan, M.J. Bahoura, Norfolk State University

The need for efficient energy utilization is driving research into ways to harvest waste-heat which is ubiquitous, abundant and free. Thermal harvesting is a promising method for capturing freely available heat and converting it to a more usable form, such as electrical energy. Thermal harvesting for low power electronic devices using ferroelectric materials is one of the emerging areas of research because they possess spontaneous polarization and exhibit excellent piezoelectric as well as excellent pyroelectric coefficients. These materials are unique as they only sense time-dependent temperature change to generate electric power. We have grown lead-free BaZr_{0.2}Ti_{0.8}O₃ (BZT)/Ba_{0.7}Ca_{0.3}TiO₃ (BCT) multilayer heterostructures and studied the structural, dielectric, ferroelectric, pyroelectric and energy density characteristics. The BZT/BCT multilayer epitaxial heterostructures were grown on SrRuO (SRO) buffered SrTiO (STO) single crystal substrate by optimized pulsed laser deposition technique. The large angle x-ray scans showed only diffraction peaks from the substrate and pseudocubic reflections (00l) from the multilayer heterostructure, confirming that these films are phase pure and epitaxial in nature. The atomic force microscopy (AFM) studies indicate that the surface roughness is low and that film growth is of high quality. The ferroelectric phase transitions have been probed above room temperature

with relaxor behavior. The polarization versus electric field (P-E) measurement exhibits well-saturated hysteresis loop with maximum and remnant polarization of 138 and 64 μC/cm², respectively. Solid-state, thin-film devices, that convert low-grade heat into electrical energy, are demonstrated using pyroelectric Ericsson cycles, and their performance is optimized by independently enhancing pyroelectric coefficient and suppressing dielectric permittivity in compositionally graded heterostructures. Our findings suggest that pyroelectric devices may be competitive with thermoelectric devices for low-grade thermal harvesting.

11:20am **TF+EM+NS+SS-ThM11 Thermal Treatment Effects on the Thermoelectric Devices from Sn/Sn+SnO₂ Thin Films**, *Satilmis Budak*, E. McGhee, Z. Xiao, E. Barnes, R. Norwood, Alabama A&M University

Approximately two-thirds of energy is lost as waste heat; the direct harvest of this waste heat using thermoelectric (TE) materials has attracted worldwide interest. TE materials can convert waste heat from industrial processes, furnaces, and engine exhaust streams into useful electricity by the Seebeck effect. The energy conversion efficiency is shown by the dimensionless figure of merit, ZT, and $ZT=S^2\sigma T/K$, where S is the Seebeck coefficient, σ is the electrical conductivity, K is the total thermal conductivity, and T is the absolute temperature. The numerator $S^2\sigma$ defines the power factor (PF), which primarily relates to the electric properties [1]. When operating as an energy-generating device, the TE device is termed a thermoelectric generator (TEG). The source of thermal energy manifests itself as a temperature difference across the TEG. When operating in a cooling or heating mode the TE device is termed a thermoelectric cooler (TEC). Similarly, the TE device produces heating or cooling that takes the form a heat flux which then induces a temperature difference across the TEC. TE devices are solid-state mechanisms that are capable of producing these three effects without any intermediary fluids or processes. For power generation applications TE devices are used in automobiles as exhaust gas waste heat recovery devices where thermal energy is scavenged along the exhaust line of a vehicle and converted into useful electricity [2]. The TE devices from 50 alternating layers of Sn/Sn+SnO₂ thin films were prepared using DC/RF Magnetron Sputtering. They were heat treated at different temperatures to form nanostructures to increase the Seebeck coefficients and electrical conductivity and decrease thermal conductivity. Seebeck coefficient, van der Pauw resistivity, and thermal conductivity were used for the characterization. SEM/EDS was used to characterize the surface morphology of the films.

[1] Hongchao Wang, Wenbin Su, Jian Liu, Chunlei Wang, "Recent development of n-type perovskite thermoelectrics", *J Materomics* 2 (2016) 225-236

[2] Chetan Jangonda, Ketan Patil, Avinash Kinikar, Raviraj Bhokare, M.D.Gavali, "Review of Various Application of Thermoelectric Module", *International Journal of Innovative Research in Science, Engineering and Technology* Vol. 5, Issue 3, (March 2016), 3393-3400.

Acknowledgement

Research was sponsored by NSF with grant numbers NSF-HBCU-RISE-1546965, NSF-EPSCOR-R-II-3-EPS-1158862, NSF-MRI-1337616, DOD with grant numbers W911 NF-08-1-0425, and W911NF-12-1-0063, U.S. Department of Energy National Nuclear Security Administration (DOE-NNSA) with grant numbers DE-NA0001896 and DE-NA0002687.

11:40am **TF+EM+NS+SS-ThM12 Thermoelectric Properties of Efficient Thermoelectric Devices from Sb/Sb+SnO₂ Thin Films**, *Eshirdanya McGhee*, S. Budak, Z. Xiao, N. Caver, B. McNeal, Alabama A&M University

The thermoelectric (TE) concept could be seen as a perfect solution for recovering waste heat from engine exhaust and converts it to electric energy. TE generators are all solid-state devices that convert heat into electricity. Unlike traditional dynamic heat engines, TE generators contain no moving parts and are completely silent. Such generators have been used reliably for over 30 years of maintenance-free operation in deep space probes such as the Voyager missions of NASA. TE systems can be easily designed to operate with small heat sources and small temperature differences [1]. An ideal TE material behaves like an electron crystal and phonon glass, allowing a large temperature gradient across it while conducting electricity efficiently to generate a TE voltage. Significant progress in the TE performance of materials has been made by exploring ultra low thermal conductivity at high temperature and reducing thermal conductivity by nano-structuring, as well as by resonant doping and energy-dependent scattering of electrons [2]. The figure of merit ZT describes material performance. ZT depends on the thermoelectric material properties of Seebeck coefficient S, electrical conductivity σ , and thermal conductivity K, and $ZT=S^2\sigma T/K$ where T is the temperature of the material

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[3]. TE devices from 50 alternating layers of Sb/Sb+SnO₂ thin films were prepared by DC/RF Magnetron Sputtering. TE devices were annealed at different temperatures to form nano-structures to increase the Seebeck coefficients and electrical conductivity and decrease thermal conductivity. For the characterization, Seebeck coefficient, van der Pauw resistivity, and thermal conductivity were used. The surface morphology was characterized using SEM/EDS.

[1] Krishna Purohit, Sheetal Kumar Jain, Dr. P M Meena, Khushaboo Singh, Manish Dadhich,

“Review Paper on Optimizations of Thermoelectric System”, International Journal of Innovative Research in Engineering & Management (IJIREM), ISSN: 2350-0557, Volume-3, Issue-4, (July-2016), 259-263.

[2] Kedar Hippalgaonkar, Ying Wang, Yu Ye, Diana Y. Qiu, Hanyu Zhu, Yuan Wang, Joel Moore, Steven G. Louie, and Xiang Zhang, “High thermoelectric power factor in two-dimensional crystals of MoS₂”, PHYSICAL REVIEW B 95, 115407 (2017) 1-9.

[3] Saniya LeBlanc, Sustainable Materials and Technologies 1–2 (2014) 26–35.

Acknowledgement

Research was sponsored by NSF with grant numbers NSF-HBCU-RISE-1546965, NSF-EPSCOR-R-II-3-EPS-1158862, NSF-MRI-1337616, DOD with grant numbers W911 NF-08-1-0425, and W911NF-12-1-0063, U.S. Department of Energy National Nuclear Security Administration (DOE-NNSA) with grant numbers DE-NA0001896 and DE-NA0002687.

12:00pm **TF+EM+NS+SS-ThM13 3D Printed Triboelectric Nanogenerator**, *I. Fattah, E. Utterback, Naga Srinivas Korivi, V. Rangari*, Tuskegee University

We report on the development of polymer nanocomposite layers made by 3D printing. The nanocomposite is composed of polydimethylsiloxane (PDMS), barium titanate nanoparticles, and multi-walled carbon nanotubes. Flexible layers of this composite have been 3D printed using a commercial 3D printer, and function as triboelectric energy generators. To the best of our knowledge, this is the first report of a PDMS based triboelectric nanogenerator fabricated by 3D printing. The nanogenerators have been evaluated in contact and separation mode and produce a maximum of 2.6 Volts under pressure from a human finger.

The fabrication procedure involves sonicating barium titanate (BaTiO₃, Skyspring Nanomaterials) and multi walled carbon nanotubes (MWCNT, Skyspring Nanomaterials) together in ethyl alcohol. This is followed by removing the excess ethyl alcohol, and manually grinding the nanoparticle powder to break any clusters. This is followed by mechanically blending liquid PDMS pre-polymer and its curing agent (~10:1 ratio by weight) with the nanoparticle powder in one beaker. Finally, the blend is filled into a dual plastic syringe, which is loaded onto an extrusion printing head of a commercial 3D printer (Hydra 16A, Hyrel LLC, USA). The printer reads a software file that defines the pattern or shape to be printed and dispenses the material from the syringe accordingly onto a base plate. For printing this composite, the base plate temperature was maintained between 75 – 90 °C, to allow curing within a few minutes. Once cured, the solid composite layers (270 μm thickness) can be peeled off the base plate.

The 3D printed PDMS-BaTiO₃-MWCNT layers have been evaluated as triboelectric energy generation. In one embodiment, the 3D printed functions as the negatively charged layer in a contact-separation scheme. A polyimide sheet is used as positively charged layer. Carbon tapes are used as current collectors on both positive and negative charged layers. When these two layers are brought in contact with some pressure applied by a human finger, and then released, characteristic negative and positive voltage spikes are respectively observed. Peak voltages as high as 2.6 Volts have been obtained with the present 3D printed PDMS-BaTiO₃-MWCNT layers. These observations indicate the applicability of this 3D printed composite in triboelectric energy generation.

Acknowledgments: This research was supported by the National Science Foundation grant #1827690.

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