

Wednesday Morning, November 12, 2014

Accelerating Materials Discovery for Global Competitiveness Focus Topic
Room: 302 - Session MG-WeM

Design of New Materials

Moderator: Christian Elsässer, Fraunhofer Institute for Mechanics of Materials IWM, Susan Sinnott, University of Florida

8:20am **MG-WeM2 Hydrogen Molecules Distribution in Multi-Cathodes Funneling Gun**, *Erdong Wang, I. Ben-Zvi, J. Skaritka, T. Rao*, Brookhaven National Laboratory, *R. Bothell, J. Bothell, A. Henry*, Atlas Technologies

The high average current polarized electron source for future electron ion collider (EIC) requires extremely high vacuum. Currently, we have constructed a DC gun based on principle of beams funneling and this gun is under commissioning. Superlattice GaAs cathodes will be used as our electron source. The lifetime of these cathodes is dependent on ion back bombardment which caused by the residual gas pressure. Multiple GaAs cathodes with almost same lifetime are extremely crucial for the operation of this gun. Therefore, produce a uniform extremely high vacuum environment is essential in gun design. We studied residual molecules distribution in funneling gun by Molflow+ and a python based Monte-Carlo simulator. Twenty cathodes testing is under planning. Cathodes lifetime changes will give us indication on vacuum distribution inside the gun. This articles describes our coding and modeling for the gun vacuum, analyzes the residual gas distribution on the gun and discusses on multiple cathodes lifetime measurement test. We also discusses the XHV achievement in our gun test.

8:40am **MG-WeM3 Tailored Functionality of Wide Band Gap Semiconductors**, *B.E. Gaddy, Z.A. Bryan, I.S. Bryan, R. Kirste*, North Carolina State University, *J. Xie, R. Dalmau, B. Moody*, Hexatech Inc., *Y. Kumagai*, Tokyo University of Agriculture and Technology, Japan, *T. Nagashima, Y. Kubota, T. Kinoshita*, Tokuyama Corporation, Japan, *A. Koukitu*, Tokyo University of Agriculture and Technology, Japan, *R. Collazo, Z. Sitar, Douglas Irving*, North Carolina State University

Semiconductors obtain new functionality by the incorporation of dilute concentrations of impurities. This principle has been utilized to tailor the properties of narrow gap semiconductors, such as Si or GaAs, but tailoring the properties of wide band gap materials for optical or high power applications has been extremely challenging. As a result, there has been slow progress in identifying *new material* combinations (bulk material + dilute dopants) to enable *novel functionality*. To help overcome these obstacles, we have developed and implemented a point defects database in which we store formation, ionization, and optical transition energies determined by hybrid exchange-correlation density functional theory methods. These predictions from first principles are then strongly coupled with synthesis and characterization efforts to identify troublesome point defects and also suggest routes to realizing desired properties if particular defect cannot be removed. The stored data is also used to solve mass balance equations to determine the number of carriers and compensating defects. In this talk, I will present our recent efforts in applying these tools to determine solutions for unwanted optical absorption in AlN grown by PVT. With our data we have demonstrated that substitutional carbon on the nitrogen site is a deep acceptor. When ionized it is the source of unwanted optical absorption. This was confirmed by PL, SIMS, and HVPE. Solution of mass balance equations reveals that the compensating defect is a singly ionized nitrogen vacancy for relevant growth conditions. The presence of this defect has been confirmed by an optical emission predicted to originate from a donor acceptor pair recombination and measured by PLE spectroscopy. When carbon cannot be removed from the growth process, we have used our database to determine mechanisms important to removing unwanted optical absorption in high carbon samples. In total, this approach has accelerated the design of new materials and also led to deeper understanding of the important mechanisms, which will impact future efforts to tailor properties of AlN and its alloys.

9:00am **MG-WeM4 Substitution and Strain Control of Polarization in Multifunctional Materials**, *M. Ashton, A. Chernatynskiy, Susan Sinnott*, University of Florida

Density functional theory (DFT) calculations are used to analyze the combined effects of substitution and strain on spontaneous polarization in multiferroic perovskites, such as BiFeO₃. In particular, DFT calculations are

used within the Vienna Ab-initio Simulation Package (VASP) to examine the macroscopic spontaneous polarization in BiFeO₃ systems with varying percentages of substitution and epitaxial strain. Substitutional elements considered for the A-site (Bi), B-site (Fe), and A/B co-substitution combination include Ba and La (A-site), Co, Cr, and Ni (B-site), and Ca/Mn and La/Ru (A/B site combination). In addition, strain values between +/- 2% are considered for each system. The results provide new insights into the roles of each of these factors on polarization and will be analyzed with data mining techniques to extrapolate design principles for high-T_c perovskites with controllable polarization. This work is supported by the National Science Foundation (DMR-1307840).

9:20am **MG-WeM5 Manipulation of Site Reactivity at the Au Nanoparticle – Titania Interface through Alloying: Insights from Density Functional Theory**, *Sampyo Hong, T.S. Rahman*, University of Central Florida

It has been shown that in methanol decomposition, as in CO oxidation, on titania supported Au nanoparticle, the interfacial atoms are the most reactive [1]. When we replace the Au atom, to which methoxy is bonded at the interface of Au₁₃/TiO₂(110), by other 3d (Cr, Ni, Cu) and 5d (Pt, W) atoms, we find that the dopant atoms at the interface become more cationic than the original Au atom and the relevant activation energies for a H-C bond scission of methoxy bonded to them is reduced as compared to that for methoxy bonded to the original Au atom. On the basis of these results we propose that the activity of Au/TiO₂ interface for reactions involving C-H scission of hydrocarbons would be enhanced by increasing the density of interfacial atoms with higher oxidation state than that of gold at the interface. Work supported in part by DOE Grant No. DOE-DE-FG02-07ER15842 [1] S. Hong and T. S. Rahman, *J. Am. Chem. Soc.* **2013**, *135*, 7629.

9:40am **MG-WeM6 Structural Descriptors for Hole Traps in Hydrogenated Amorphous Silicon Revealed through Machine Learning**, *Tim Mueller*, Johns Hopkins University, *E. Johlin, J.C. Grossman*, Massachusetts Institute of Technology

The discovery and design of new materials can be accelerated by the identification of simple descriptors of material properties. However the identification of the most relevant descriptors and how they relate to the properties of interest is not always obvious. We demonstrate how machine learning, in the form of genetic programming, can be used to identify relevant descriptors for predicting hole trap depths in hydrogenated nanocrystalline and amorphous silicon. Amorphous silicon is an inexpensive and flexible photovoltaic material, but its efficiency is limited by low hole mobility. We have evaluated 243 structural descriptors of amorphous silicon to identify those that are most indicative of the hole trap depth. Our calculations reveal three general classes of structural features that influence hole trap depth and predict that multiple interacting defects may result in deeper traps than isolated defects. These results suggest a possible mechanism for the Staebler-Wronski effect, in which exposure to light degrades the performance of amorphous silicon over time.

11:00am **MG-WeM10 Integration of Meso-scale Microstructural Modeling for Engineering Materials Development**, *Veena Tikare*, Sandia National Laboratories

INVITED

The meso-scale materials modeling community, over the last two decades, has developed vast capabilities in microstructurally-based modeling of complex ceramic and metals. While several modeling techniques have been developed, the Potts kinetic Monte Carlo model and the phase-field model form the foundation of most materials modeling efforts for a variety of microstructural evolution processes experienced by ceramics and metals during fabrication and engineering service. Harnessing these modeling capabilities and applying them to design materials to tailor their microstructure for optimal engineering performance properties and designing fabrication processes to obtain the desired microstructure can greatly accelerate development and optimizing of materials for a large number of technologies. This presentation will give a general overview of the core capabilities of the microstructural evolution modeling capabilities by reviewing the two most commonly used methods, Potts and phase-field. The former is a discrete, statistical-mechanical model that has been successfully used to simulate many microstructural evolution processes, such as grain growth, sintering, coarsening in the presence of mobile and immobile pinning phase, and recrystallization. The phase-field model is a continuum, thermodynamic model that has been used to successful simulation solidification, phase transformation and coarsening processes. The capabilities and limitation of each model will be reviewed and appropriate application of the models to different materials microstructural evolution processes will be discussed. The presentation will also

demonstrate how these two models can be applied to understand and predict materials processes including coarsening, sintering and phase transformations. Specific examples of microstructural modeling and their application to design materials microstructure for optimal performance will be presented and discussed. Examples will include simulation of microstructural evolution during sintering of complex powder compacts; the generation, transport and release of fission gases from nuclear fuels during service in a reactor; and development of grain shapes and sizes during welding processes. Finally, the current trends in microstructure model development will be discussed.

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

11:40am **MG-WeM12 Insights on the CO₂ Reduction Mechanism on Bio-inspired Iron Sulphide**, *Alberto Roldan, N.H. de Leeuw*, University College London, UK **INVITED**

Carbon dioxide capture and utilisation is gaining attention, driven not only by environmental factors but also by the potential to use it as chemical feedstock. One plausible utilisation route is its conversion to form small organic molecules, however, CO₂ is thermodynamically very stable and its reduction is energy intensive. The CO₂ conversion takes place under mild conditions in chemoautotrophic bacteria catalysed by enzymes.¹ These enzymes often contain Fe₄S₄ clusters (cubanes), which have been shown to act as electron-transfer sites^{2,3} but they can also be catalytically active centres for molecule transformations.⁴ An iron thiospinel mineral is structurally similar to the cubane,⁵ fact that brings us to suggest it as a novel heterogeneous catalyst. We present a theoretical investigation using the iron sulphide greigite mineral (Fe₃S₄) as a catalyst to transform CO₂ into small organic molecules. In agreement with the experiments, the adsorbed species depends on the solution pH as well as both concentration and actual products formed. The reduction consists of a sequential hydrogenation steps that we studied by the common Langmuir-Hinshelwood mechanism. We have identified ~170 steady states describing the different reaction pathways where the most favourable ones lead to formate and carboxyl intermediates yielding two different products, HCOOH and CH₃OH, also identified experimentally under conditions of room temperature and pressure.

References

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- (3) Hayashi, T.; Stuchebrukhov, A. *Abstracts of Papers of the American Chemical Society* 2012, 243.
- (4) Seino, H.; Hidai, M. *Chemical Science* 2011, 2, 847.
- (5) Martin, W.; Russell, M. J. *Philosophical Transactions of the Royal Society of London Series B-Biological Sciences* 2003, 358, 59.

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