2011 NIST Diffusion Workshop

held in conjunction with the TMS 2011 Annual Meeting
March 3, 2011
San Diego, CA

"Addressing Phenomenological Diffusion Problems with the Lattice Monte Carlo Method," Graeme E. Murch, Irina V. Belova and Thomas Fiedler, The University of Newcastle, Australia

Prof. Graeme Murch presented his work on using the Lattice Monte Carlo method to address a variety of different diffusion problems. Munch explained that the LMC approach maps a phenomenological mass (or thermal) diffusion problem onto a very fine-grained lattice, which is then explored by random walking mass or energy 'packets'. Munch utilizes the LMC approach to investigate various grain boundary and interdiffusion problems. The LMC approach was applied to thermal diffusion problems in layered Al/paraffin composites and cellular metals.

References:

The Lattice Monte Carlo Method for Solving Phenomenological Mass and Heat Transport Problems
Irina V. Belova, Graeme E. Murch, Thomas Fiedler and Andreas Öchsner
Diffusion Fundamentals **4** (2007) 15.1 - 15.23

"The role of crystal structure and configurational disorder on ion transport in Li-electrode materials," Anton Van der Ven, University of Michigan, Ann Arbor, MI

Prof. Anton Van der Ven demonstrated how to calculate kinetic coefficients for non-dilute multicomponent solids using first principles. These calculations must address two different time-dependent processes: the short time scale process over which individual atomic hops occur and a longer time scale process over which the redistribution of atoms across the concentration gradient occurs. The individual atomic hops can be modeled with transition state theory. Statistical mechanics can be used to describe the collective phenomena of successive atomic hops that lead to redistribution over a given concentration gradient. Within this framework kinetic Monte Carlo simulations are used to evaluate Kubo-Green expressions to determine multicomponent diffusion coefficients for a variety of electrodes used in Li ion batteries.

References:

Van der Ven, A., J. C. Thomas, et al. (2010). "Linking the electronic structure of solids to their thermodynamic and kinetic properties." Mathematics and Computers in Simulation **80**(8): 1393-1410.
http://dx.doi.org/10.1016/j.matcom.2009.08.008

Van der Ven, A., H.-C. Yu, et al. (2010). "Vacancy mediated substitutional diffusion in binary crystalline solids." Progress in Materials Science **55** (8) 61-105.
http://dx.doi.org/10.1016/j.pmatsci.2009.08.001

Swoboda, B., A. Van der Ven, et al. (2010). "Assessing Concentration Dependence of FCC Metal Alloy Diffusion Coefficients Using Kinetic Monte Carlo." Journal of Phase Equilibria and Diffusion **31**(3): 250-259.
http://dx.doi.org/10.1007/s11669-010-9706-8

"Silver diffusion in copper bi- and tri-crystals,"

Sergiy V. C, Institute of Materials Physics, University of Münster, Germany

Prof. Divinski presented work on Ag diffusion in Cu bi-crystal and tri-crystals. Divinski emphasized that a better understanding of grain boundary diffusion mechanisms is essential for understanding highly deformed materials.

The relationship between microscopic mechanisms of grain boundary diffusion and grain boundary structure is of fundamental as well as engineering importance. Its solution requires, in particular, the determination of diffusion coefficients for single grain boundaries with precisely characterized structures, i.e. with specified misorientation and inclination parameters.

An overview of recent advances in experimental investigation of grain boundary diffusion and segregation in bi-and tri-crystals is presented. The radiotracer technique in combination with precise parallel sectioning by microtome was used. For the first time, diffusion of a solute (^{110m}Ag) is measured along a single grain boundary in Cu Σ5 bicrystal and along a triple line in copper tricrystal in the true dilute limit conditions. The measurements are performed under formal conditions of the C-type kinetic regime of grain boundary diffusion according to common Harrison classification. The product of the triple line diffusion coefficient, *D*tj, the triple line cross-section, *A*, and the segregation factor, *s*, was determined. The Ag diffusion rate along a general triple line is estimated to be faster than that along general high-angle grain boundaries by about three orders of magnitude at 575K in copper. Based on the diffusion data, the triple line energy is estimated.

References:
Edelhoff, H., S. I. Prokofjev, et al. (2011). "The C-regime measurements of grain boundary diffusion of silver in copper [Sigma]5 (310) bicrystal." Scripta Materialia **64**(5): 374-377.
http://dx.doi.org/10.1016/j.scriptamat.2010.10.032

Divinski, S. V., G. Reglitz, et al. (2010). "Grain boundary self-diffusion in polycrystalline nickel of different purity levels." Acta Materialia **58**(2): 386-395. http://dx.doi.org/10.1016/j.actamat.2009.09.015

"First-principles modeling of diffusion in bcc metals,"

Nils Sandberg, Royal Institute of Technology, Stockholm Sweden.

First principle calculations were performed to improve the understanding of the non-Arrhenius behavior at low temperatures. The modeling is based on large-scale DFT (Density-functional theory) MD (molecular dynamics) simulations, which used the AM05 exchange correlation functions (Mattsson PRB 72 2005). These calculations allowed both the vacancy formation and migration energies to be evaluated as functions of temperature.

Using DFT-MD simulations, the vacancy-formation energy was determined by comparing the average the thermal energies of a system with a vacancy to the bulk system, assuming a fixed volume corresponding to the relaxed state at 0 K. The enthalpy of vacancy formation increased anharmonically with temperature.

The vacancy migration rate was determined by estimating the mean jump rate of a single vacancy at 2600 K and 2800 K using DFT-MD simulations.

Reference:
Mattsson, T. R., N. Sandberg, et al. (2009). "Quantifying the anomalous self-diffusion in molybdenum with first-principles simulations." Physical Review B **80**(22): 224104.
http://dx.doi.org/10.1103/PhysRevB.80.224104

"Using First Principles to Calculate Self –Diffusion Coefficients," Prof. Zi-Kui Liu

Improved first principle diffusion coefficients for hcp Mg and Ti were presented. An 8-frequency model was used to calculate the impurity diffusion of Al, Zn, Sn and Ca on a hcp Mg lattice. The self-diffusion of hcp Ti diffusion was calculated using the projector augmented wave (PAW) method, and the exchange-correlation approximation (GGA) of Perdew–Burke–Ernzerhof (PBE).

References
Ganeshan, S., L. G. Hector Jr, et al. (2011). "First-principles calculations of impurity diffusion coefficients in dilute Mg alloys using the 8-frequency model." Acta Materialia **59**(8): 3214-3228.
http://dx.doi.org/10.1016/j.actamat.2011.01.062

Shang, S. L., L. G. Hector, et al. (2011). "Anomalous energy pathway of vacancy migration and self-diffusion in hcp Ti." Physical Review B **83**(22): 224104.
http://dx.doi.org/10.1103/PhysRevB.83.224104

Reference Self-Diffusion Discussion

The group reviewed the self-diffusion assessments for Al, Cu, Cr, Fe, and Ni and agreed on a set of reference self-diffusion mobilities for these data. The group also agreed that for anomalous diffusers (those elements that do not follow an Arrhenius relation at low temperatures), like bcc- Mo, a two-exponential function should be used instead of multiple temperature ranges. The group agreed to continue to establish reference self-diffusion mobilities. The next elements to review are Co, Mo, W, Pt, Pd, Nb, Ta, and Li (in the stable phases). The group will also work on establishing a consistent approach to defining the metastable end-member self-diffusion terms (e.g. fcc-W).

Actions Items:

- Carrie agreed to draft a report on the initial reference self-diffusion mobilities (Al, Cu, Cr, Fe and Ni).
- Henrik agreed to check how well the current bcc-Fe description compared to Nils first principle results at low temperature.