

## Catalysis Club of Philadelphia

Thursday March 19<sup>th</sup>, 2015

DoubleTree Hotel

4727 Concord Pike Wilmington, DE 19803

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### Density Functional Theory Investigation of Hydrogenation and Dehydrogenation Reaction on Binary Metal Alloys: Effect of Surface Ensembles and Composition

Fuat E Celik

*Department of Chemical and Biochemical Engineering  
Rutgers, The State University of New Jersey, Piscataway, NJ 08854*

**Social Hour: 5:30 PM**

**Dinner: 6:30 PM**

**Meeting: 7:30 PM**

**Members: \$35.00**

**Walk Ins & Non-members: \$40.00**

**Student & Retired Members: \$20.00**

### Menu

**Seared Yellow Fin Tuna** – cucumber wasabi-teriyaki glaze, sautéed spinach with jasmine rice;

**Rosemary Dijon Chicken** – sautéed chicken breast with roasted cherry tomatoes and a rosemary Dijon cream sauce served with her roasted potatoes and fresh green beans

**Grilled Vegetable Ravioli** – grilled vegetables blended with ricotta, mozzarella, parmesan, fontina & pecorino romano cheese wrapped in basil pasta;

**Meal reservations** - Please notify your company representative or Eyass Mahmoud (eyass@udel.edu) by **Thursday March 12<sup>th</sup>**, or register online:

<http://catalysisclubphilly.org/program/meeting-registration/>

Company Representatives – We would like to encourage you to make meal/meeting reservations to your company representative.

**Membership** - Dues for the 2014-15 season will be \$25.00 (\$5.00 for the local chapter and \$20.00 for the national club). Dues for students, post-docs and retirees will be \$10.00 (\$5.00 for local club and \$5.00 for national club).

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**Abstract.** In supported metal catalysts, the tradeoff between activity and selectivity presents an important challenge for catalyst design. By allowing two dissimilar metals, we can attempt to tune the selectivity of the catalyst by enhancing bond-formation and desorption rates through the addition of a less-reactive element, while maintain high bond dissociation activity from the more active metal. The resulting catalyst properties depend strongly on the catalyst composition and ratio of the two metals (electronic effect), but may also depend on the local structure of surface ensembles of the alloy components (geometric effect). In this talk we will explore two examples of binary alloys where surface composition and geometry play an important role in determining the selectivity of the catalyst through density functional theory (DFT).

In the first example, we have examined the effect of platinum tin alloy structure and composition on the kinetics and thermodynamics of dehydrogenation and coke formation pathways during light alkane dehydrogenation. Light alkane dehydrogenation to olefins can add significant value to hydrocarbon processes that generate ethane and propane by converting low value commodity fuels to high-value chemical and polymer precursors. Supported Pt catalysts are known to be active but show significant coke formation and deactivation, which can be alleviated by alloying with Sn and other main group elements. We aim to understand how the structure and composition of these alloys affect their ability to suppress coke formation. We investigate the potential energy surfaces from ethane along the desired pathway to ethene, and along the undesired pathways towards surface carbon/coke. The effect of Pt/Sn ratio and surface geometry is investigated. As compared to pure Pt, bond scission is more difficult on the alloys and desorption is more facile, and both effects are enhanced as three-fold hollow sites consisting of only Pt atoms are eliminated.

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In the second example, we evaluate Au/Ni near-surface alloys as potential oxygen reduction catalysts for the direct synthesis of hydrogen peroxide from O<sub>2</sub> and H<sub>2</sub>, thereby avoiding the current anthraquinone process. While Au may have higher O-H bond formation activity, it is a poor O<sub>2</sub>-dissociation catalyst, and likewise Ni is very effective at O<sub>2</sub>-dissociation but not oxygen hydrogenation. Alloying Au with Ni(111) lowers H<sub>2</sub> dissociation barrier while keeping the O<sub>2</sub> dissociation barrier large relative to O<sub>2</sub> hydrogenation. Desorption of H<sub>2</sub>O<sub>2</sub> is similarly competitive with H<sub>2</sub>O<sub>2</sub> dissociation on alloy surfaces. However, the selectivity for the OOH radical remains a challenge, with barrierless O-O bond dissociation and large (1.3 eV) hydrogenation barriers. We further investigate how the Au/Ni surface may rearrange itself to regenerate three-fold hollows of Ni atoms in the presence of strongly adsorbing surface species.

