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Project Title: Study of Petroleum Related Heterogeneous Catalysis via Correlated Fluorescence and Electron Microscopy

Emilie Ringe, PhD, Materials Science and NanoEngineering, William Marsh Rice University, Houston, TX

Since the start of this grant (Sept. 1, 2016) the PI, Dr. Ringe, has led a team of 2 undergraduate students, four graduate students (two supported by NSF), and one postdoctoral associates in a quest for developing new catalysts and new means to observe catalytic reactions. The progress made on this project, as well as the contribution made to other catalysis projects are described here.

In addition to the scientific progress made possible by the financial support of ACS-PRF, the PI reports here on several career contributions. First, two of the graduate students involved graduated from Rice with a number of research papers and went on to stable positions, one in the scientific industry and one in a support role in academia. The postdoctoral student supported by this grant enjoyed much flexibility and landed a research associate professor position at UC Boulder. Finally, the recognition and financial support of ACS-PRF supported the PI's application to a position at a top 5 institution in the world, and is now a lecturer (tenured assistant professor) at the University of Cambridge. The support of the ACS-PRF, early on in the group's existence, clearly had a major positive impact.

Several publications have resulted from the efforts concentrating on multimetallic catalytic nanoparticles:

D. F. Swearer, R. K. Leary, R. Newell, S. Yazdi, H. Robatjazi, Y. Zhang, D. Renard, P. Nordlander, P. A. Midgley, N. J. Halas,* E. Ringe,* "Transition Metal Decorated Aluminum Nanocrystals" ACS Nano (2017), 11, 10281-10288

A simple polyol synthesis is presented as a flexible route to produce aluminum nanocrystals decorated with eight varieties of size-tunable transition-metal nanoparticle islands, many of which have precedence as heterogeneous catalysts.

S. M. Rehn, E. Ringe,* "Controllably Hollow AgAu Nanoparticles via Nonaqueous, Reduction Agent-Assisted Galvanic Replacement" Part. Part. Syst. Char. (2018), 35, 1700381

Another example of a multimetallic synthesis approach to potentially catalytically active structures, with the additional opportunity to include a non-aqueous cargo.

C. Zhang, S. Yang, J. Wu, M. Liu, S. Yazdi, M. Ren, J. Sha, J. Zhong, K. Nie, A. S. Jalilov, Z. Li, H. Li, B. I. Yakobson, Q. Wu, E. Ringe, H. Xu, P. M. Ajayan, J. M. Tour,* "Electrochemical CO₂ Reduction with Atomic Iron-Dispersed on Nitrogen-Doped Graphene" Adv. Energy Mater. (2018), 1703487

A collaborative project looking at the catalytic properties of Fe-doped graphene; the PI's group provided mainly TEM support and this allowed the group to be exposed to new, interesting materials.

J. R. Daniel, L. A. McCarthy, S. Yazdi, M. Chagnot, E. Ringe,* D. Boudreau* "Gold Speciation and Co-Reduction Control the Morphology of AgAu Nanoshells in Formaldehyde-Assisted Galvanic Replacement" J. Phys. Chem. C (2018) 122, 18168-18176

Here, the pH of formation of co-reduction/galvanic replacement of Ag/Au shells was shown to dictate their morphology and surface composition. This is key to catalytic applications because the nature of the atoms present at the particle's surface

J. S. Biggins, S. Yazdi, E. Ringe,* "Magnesium Nanoparticle Plasmonics" Nano Lett. (2018) 18, 3752-3758

One of the key challenge we face in plasmon-enhanced catalysis is the cost of the plasmonic material. Therefore, toward the end of this grant, we investigated the possibility of making (and eventually using) non-noble metals for plasmonics in the visible range. This has proven highly successful and led to a

publication in Nano Letters. Further works are currently ongoing to decorate these particles and test their catalytic properties.

Up to three additional publications acknowledging support from the ACS-PRF are submitted or in preparation.