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Plasmonic Catalysis: Heating vs. Hot Electrons

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In plasmon-enhanced heterogeneous catalysis, illumination accelerates reaction rates by generating hot carriers and hot surfaces in the constituent nanostructured metals. In order to understand how photo-generated carriers enhance the non-thermal reaction rate, the effects of local heating and thermal gradients in the catalyst bed must be confidently and quantitatively characterized. This is a challenging task considering the conflating effects of light absorption, heat transport, and reaction energetics. Here, we introduce a methodology to distinguish the thermal and non-thermal contributions from plasmon-enhanced catalysts, demonstrated by illuminated rhodium nanoparticles on oxide supports to catalyze the CO_2 methanation reaction. By simultaneously measuring the total reaction rate and the temperature gradient of the catalyst bed, the effective thermal reaction rate may be extracted. The residual non-thermal rate of the plasmon-enhanced reaction is found to grow with a super-linear dependence on illumination intensity, and its apparent quantum efficiency reaches ~46% on a Rh/TiO_2 catalyst at a surface temperature of 350 °C. Heat and light are shown to work synergistically in these reactions: the higher the temperature, the higher the non-thermal efficiency in plasmon-enhanced catalysis.

Biography:

Jie Liu is currently the George B Geller Professor of Chemistry at Duke University. He earned a B.S. in Chemistry from Shandong University in 1987 and a Ph.D. in Chemistry from Harvard University in 1996. His research interests include synthesis and chemical functionalization of nanomaterials, plasmonic catalysis, nanoelectronic devices, scanning probe microscopy, and carbon nanomaterials. As a faculty member, Professor Liu has received the DuPont Young Professor Award, Outstanding Oversea Young Investigator Award from NSF-China, Ralph E. Powe Junior Faculty Enhancement Award from Oak Ridge Associated Universities, and Bass Professorship from Duke University for excellence in teaching and research. He is elected as a Fellow in AAAS (2013), APS (2014) and RSC (2013). He also serves as an associate editor for RSC journal Nanoscale since 2012.