Special Seminar in Energy Research

at the

University of Pennsylvania

Charge Transfer and Chemistry at Catalytic Interfaces

Abstract

Careful studies of catalysts under reaction conditions have revealed that the surface properties of a material cannot simply be extrapolated from a bulk crystal structure and that it is the often surprising properties of a surface or interface that give rise to its useful catalytic properties. In this talk I will describe two frontier experimental approaches that are used to study the relationship between charge transfer and chemistry at catalytic interfaces. First, sum frequency generation (SFG) vibrational spectroscopy is used to follow the progression of molecular intermediates during the furfuraldehyde hydrogenation reaction on a Pt/TiO_2 catalyst. The results of this experiment show that a charge transfer interaction on the active TiO_2 support produces an ionic reaction intermediate, and that this charged intermediate is the highly active precursor to selective C=O bond hydrogenation. In a second experiment, ultrafast x-ray spectroscopy is used to follow a chemical reaction, this time from the perspective of the charge carrier. During photocatalytic oxidation of methanol on a Co_3O_4 catalyst, I measure the effect of hole transfer on the bulk carrier lifetimes in photoexcited Co_3O_4. The results show that fast hole transfer from Co_3O_4 to methanol leads to a surface mediated carrier relaxation pathway that dominates bulk carrier lifetimes in the Co_3O_4 catalyst. This combination of approaches allows observation of charge transfer reactions from the perspective of molecular intermediates as well as the ultrafast dynamics of charge carriers that actually drive the surface chemistry. The results of these experiments emphasize the need to characterize catalytic interfaces under relevant reaction conditions in order to understand the mechanisms of catalytic selectivity and energy conversion efficiency.

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