601a Stochastic and Dimensional Analysis of High-Pressure Hydrogen Adsorption Via Spillover on Carbon Supported Catalyst

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Carbon metal synergy in the form of hydrogen spillover can significantly increase the uptake of a carbon-based hydrogen storage material. The fundamental processes involved in hydrogen spillover are: ***adsorption*** of active hydrogen species, ***desorption*** of hydrogen species as molecular hydrogen and ***surface diffusion*** of hydrogen species. These processes cannot be decoupled for experimental measurements and thus no model for spillover has been universally adopted. Several reports suggest that the rates of these processes are system dependent and it is not fully clear whether or when the hydrogen spillover process is a function of metal dispersion, metal particle size, metal loading, and metal-carbon interface. Our hypothesis that hydrogen surface coverage will be determined by the competing rates of desorption versus surface diffusion lends itself to a dimensional analysis of the spillover process. Our dimensional analysis, combined with a kinetic Monte Carlo model, studies the effect of hydrogen pressure; relative activation energies of the processes involved; active metal and carbon surface area; and length of metal-support interface. We anticipate that these theoretically-grounded models facilitate optimization of hydrogen storage materials beyond a “trial and error” approach and will also address some of the seeming contradictions in earlier hydrogen spillover literature applied to heterogeneous catalysis.