Micro-kinetics of NO_x Storage and Reduction with $H_2/CO/C_3H_6$ on $Pt/BaO/Al_2O_3$ Monolith Catalysts

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Abstract:

Steady state NO_x storage and reduction on $Pt/BaO/Al_2O_3$ by $H_2/CO/C_3H_6$ reactant mixtures is studied using an isothermal, 1+1 dimensional two-phase model that accounts for washcoat diffusional limitations. It predicts the storage and reduction performance over a wide range of inlet feed concentrations and temperatures. Global kinetic models have a major limitation of not being able to predict accurately the performance of the catalyst with changing feed composition. To account for such variations, micro-kinetic models elucidating the role of individual reductants have been developed. The temperature dependence of the rate constants has been quantified to reflect the surface chemistry that is consistent with experimental observations. Figure 1 shows the comparison between the experimental and model predicted data for NO/H_2, NO/CO and NO/C_3H_6 systems. The model predicts the experimentally observed trends in individual systems reasonably well and is further used to predict the conversions and selectivities for a combined $NO/H_2/CO/C_3H_6$ system.

Inclusion of washcoat diffusion enables the model to accurately predict the NO_x conversion and scale-up performance of the catalyst over a wide temperature range. Figure 2 shows that the model accurately represents different controlling regimes during NO/H_2 reaction. It can be seen that the system goes from being kinetic controlled to washcoat diffusion controlled as temperature increases. At higher temperatures most of the reaction is restricted to a thin layer of the washcoat close to

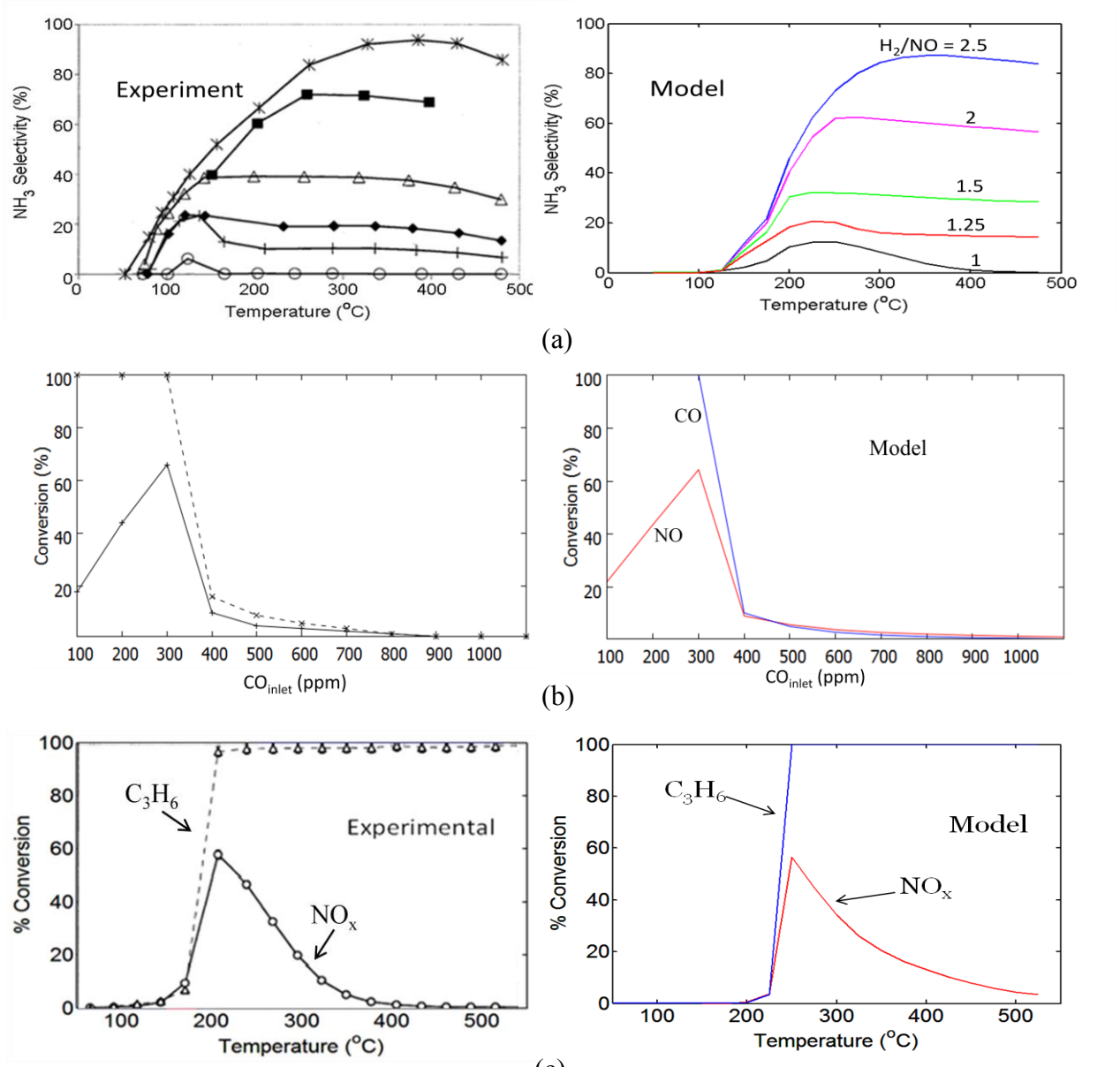


Figure 1: Comparison of the experimental and model predicted conversions and selectivities (a) NH_3 selectivity in NO/H_2 system (b) NO and CO conversions in NO/CO system and (c) NO and C_3H_6 conversions in NO/C_3H_6 system.

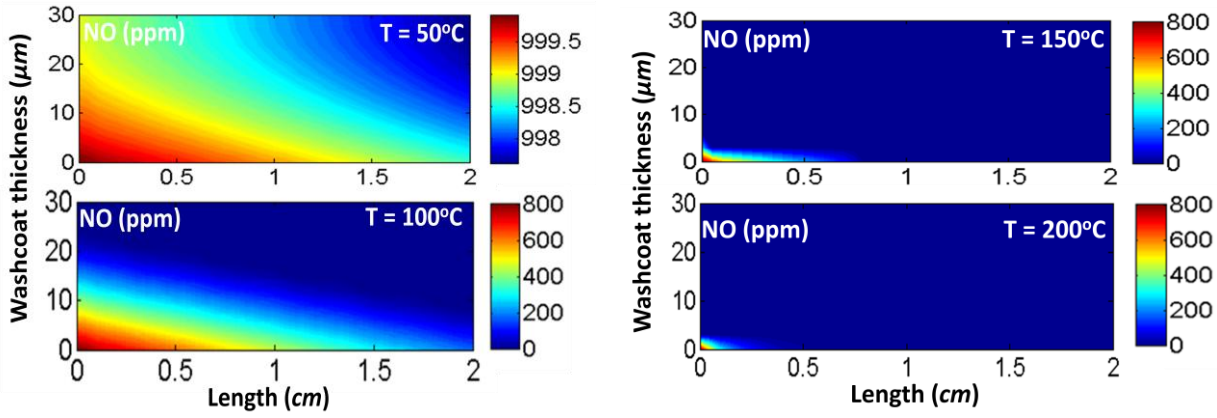


Figure 2: NO concentration inside the washcoat at different temperatures for NO/H_2 system [NO_{inlet}=1000ppm, H_{2inlet}=2500ppm; Pt loading = 2.2 wt%].

the fluid-washcoat interface. The sensitivity of the estimated kinetic parameters to the effective diffusivities in the washcoat is also analyzed by pore scale comparison between the time constants of adsorption ($C_0/R_{ads}(C_0)$) for $H_2/CO/C_3H_6$ and pore scale diffusion time constants ($\frac{r_{eff}^2}{D_{eff}}$). Results indicate that the estimated micro-kinetic parameters are influenced by the washcoat pore structure and the properties of the precious metal and storage components.