Development of a kinetic model to evaluate water storage during cold start on commercial Cu-SAPO SCR catalysts

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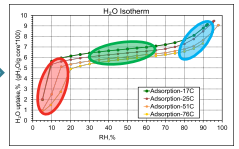
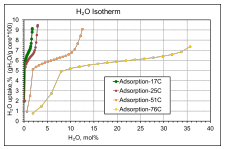
Motivation

- Develop a kinetic modeling approach to evaluate amount of water storage during cold start on commercial Cu SAPO-34 catalysts
- Design well-planned Transient TPD and steady state isotherms testing to understand water adsorption and desorption characteristics
- Utilize kinetic model to simulate water storage dynamics and provide control strategy insight for cold start operation

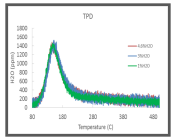
Experimental Investigation

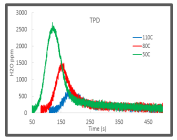
- Steady state H_2O Isotherms: Monolith sample (40-90 mg) was saturated with H_2O for 4 hrs after pretreatment at 350C for 30 mins
  - Gravimetric water adsorption analyzer is used to determine H_2O isotherms at various steady state temperatures and Relative Humidity (RH)
  - Water uptake by wt % increases with concentration in three regimes: (red) Initial fast uptake (green) slow uptake (blue) bulk condensation
  - Water storage decreases sharply with temperature

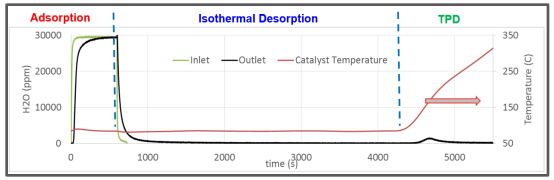




- Transient TPD: Identify time required to saturate the catalyst with H_2O and rate of H_2O removal
  - SCR catalyst is saturated by flowing H_2O for sufficient duration at 80C (Adsorption) followed by Isothermal desorption to measure weakly adsorbed H_2O and, finally TPD
  - Unique set up: Introduced high amount of H_2O at Cu-SAPO 34 micro core inlet by oxidation of H_2 and O_2 over DOC
  - Total H_2O adsorption and isothermal desorption increases with H_2O concentration
  - TPD at multiple inlet water concentrations showed that strongly bounded water is nearly independent of feed concentration
  - Total H_2O adsorption and desorption decreases with increase in adsorption temperature







Kinetic Model & Modeling Results

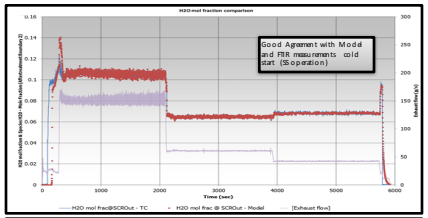
- 2-site model was developed with strong and weak bonded sites to release water based on SCR bed temperature

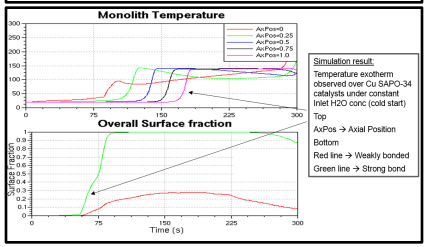
  Rate of adsorption= $k_{ads}C_{H2O}(1-\theta)$

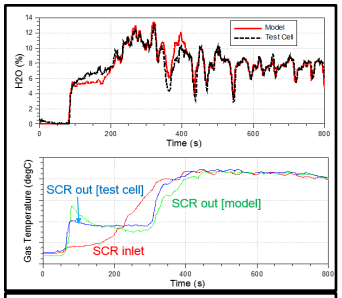
  Rate of desorption =$k_{des}\theta$

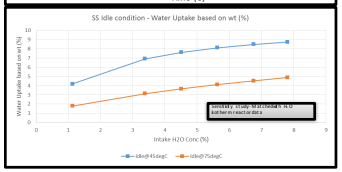
  where $k_{des} = A_{des}exp\left[\frac{-E_{des}}{RT}(1-\alpha\theta)\right]$

- Site densities for maximum possible water storage were calculated based on experimental data
- Water adsorption step over SAPO zeolite catalyst is an exothermic reaction
  This observed phenomenon from experimental data and literature studies was captured as a modeling criteria[1]
- During temperature ramp up, water desorption is accompanied with temperature drop due to an endothermic reaction
- Model predicts axial distribution of solid temperature based on water adsorption/ desorption events
- Water storage estimation across Cu-SAPO-34 catalysts showed good agreement under cold start, transient (FTP) cycles and steady state cycles Test Cell conditions
- Sensitivity study has been performed at series inlet temperatures and water concentrations. Compared with reactor data









Summary

- Extension of existing literature studies and in-house experimental results were captured through modeling efforts
- Verification and validation for the Water storage model was performed through series of Steady state and transient field testing
- Developed state of art model had good agreement predicting water storage and provides tuning strategy insight on cold start emissions

Reference:
1. Schmeisser et al., SAE Int. J. Commer. Veh. 6(1):2013