Kinetic modeling of H\textsubscript{2}-assisted C\textsubscript{3}H\textsubscript{6} Selective Catalytic Reduction of NO over Silver Alumina catalyst

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Introduction

\begin{itemize}
  \item Ag/Al\textsubscript{2}O\textsubscript{3} catalyst is a potential catalyst for NO\textsubscript{2} abatement with HC-SCR technology. Further, addition of H\textsubscript{2} has been shown to give better HC-SCR performance on Ag/Al\textsubscript{2}O\textsubscript{3}[1].
  \item The present work aims to set up a global kinetic model to describe mechanistic role of H\textsubscript{2} to promote C\textsubscript{3}H\textsubscript{6}SCR over Ag/Al\textsubscript{2}O\textsubscript{3} catalyst.
\end{itemize}

Experimental methods

\begin{itemize}
  \item Ag/Al\textsubscript{2}O\textsubscript{3} with 2 wt.% Ag loading was prepared by sol-gel freeze dried method. The catalyst powder was washcoated on cordierite monolith.
  \item Activity measurement was conducted in a flow reactor equipped with gas FTIR and MS.
  \item Temperature-programmed reaction and transient experiments were used.
  \item The model aimed to simulate wide range of temperature and inlet feed concentration.
\end{itemize}

Modeling methods

Development of reaction mechanisms:

\begin{itemize}
  \item Experimental data shows NO oxidation and C\textsubscript{3}H\textsubscript{6} SCR are always higher in the presence of H\textsubscript{2} even for high temperature range.
  \item Initial screening of mechanisms suggested that single role of H\textsubscript{2} to remove inhibiting nitrate was insufficient to reproduce wide range of experimental conditions.
  \item Key point in reaction mechanism: H\textsubscript{2} is proposed to have dual role to remove inhibiting nitrate and simultaneously form more active reduced sites, S\textsubscript{r}.
\end{itemize}

\begin{equation}
\begin{array}{c}
\text{H}_2(\text{g}) + \text{NO} + \text{H}_2\text{O} \\
\xrightarrow{\text{Al}_2\text{O}_3} \text{S}_\text{r} \quad \text{Oxidized Ag} \\
\text{C}_3\text{H}_6, \text{H}_2, \text{H}_2\text{O} \\
\end{array}
\end{equation}

- Reactor model: single channel model was used to describe mass transfer and reaction kinetic. The model also includes mass transfer of gas components inside washcoat [2].

Results and discussion

A sample of experimental and simulation results:

- The proposed H\textsubscript{2} role reproduced experimental data well for both temperature-programmed and transient experiments. Similarly, model validation under NO, C\textsubscript{3}H\textsubscript{6} oxidation and C\textsubscript{3}H\textsubscript{6} SCR with/without H\textsubscript{2} at 250 and 400\degree C gave good agreement between simulations and transient experiments.
- All reactions on reduced Ag sites (S\textsubscript{r}) are more rapid than on oxidized Ag sites (S\textsubscript{o}) which is indicated by lower activation energies on S\textsubscript{r} than on S\textsubscript{o} for the same reactions.
- Weisz modulie evaluations were made for H\textsubscript{2}-assisted C\textsubscript{3}H\textsubscript{6} SCR and C\textsubscript{3}H\textsubscript{6} SCR indicated that larger fraction of experimental data was free from mass transport resistance.

Conclusions

- A global kinetic model for H\textsubscript{2}-assisted C\textsubscript{3}H\textsubscript{6} SCR has been developed to effectively capture a wide range of feed concentrations and temperature.
- The model proposes dual role of H\textsubscript{2} to eliminate nitrate as well as to form a reduced sites, S\textsubscript{r} NO oxidation, H\textsubscript{2} oxidation as well as C\textsubscript{3}H\textsubscript{6} SCR reactions are more rapid on the reduced sites, S\textsubscript{r}.
- A larger fraction of experimental data was free from the influence of mass transport resistance.

References


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