Modeling and Experimental Investigation of Zeolite Catalysts for NH_3-SCR Aftertreatment

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Introduction

Urea-SCR catalysts are regarded as the leading NO_x aftertreatment technology to meet the 2010 NO_x emission standards for on-highway vehicles running on heavy-duty diesel engines. Unfortunately, during low temperature engine operating conditions, urea droplets might not evaporate and thermolyze to form NH_3 to reduce NO_x in diesel exhaust. Since urea breaks up into NH_3 and HNCO, there is a need to understand the reactions of both the species on an SCR catalyst. This talk presents a brief overview of modeling, system identification and validation of a Fe-zeolite catalyst for NH_3-SCR aftertreatment. Mathematical equations governing the SCR catalyst along with assumptions are explained. The model is coded as a C language S-function and the simulation is implemented in Matlab/Simulink. Steady state data on a Fe-zeolite catalyst is used to identify the parameters using Matlab's optimization toolbox and the model is validated against a set of data not used for calibration. The model predicted outputs are then analyzed for both steady state and transient conditions.

Test Procedure

To characterize the catalyst, two different steady state tests are conducted. 1. Surface isotherm tests followed by temperature programmed desorption (TPD)[1] to identify the adsorption parameters and 2. Transient response tests suggested by Malmberg et al.[2] to identify the rate parameters associated with three SCR reactions, NH_3 oxidation, NO oxidation and N_2O formation reactions. For the surface isotherms, three temperatures 200^0C, 350^0C and 450^0C are chosen along with three concentrations 100 ppm, 200 ppm and 350 ppm to yield a test matrix of 9 points. For the transient response tests, a stream of 350 ppm of NO_x, 350 ppm of NH_3, 4.5% H_2O, 4.5% CO_2, 14% O_2 and balance N_2 is used. A schematic of a typical transient response test is shown in Figure 1.

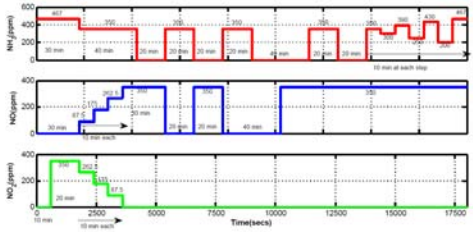

Figure 1: Transient Response Test for System Identification

To investigate the NH_3/NO_x (alpha) effect on NO_x conversion during the transient response tests, the concentrations are varied based on reaction stoichiometry.

System Identification

The identification phase involves two steps. During the first step, the adsorption/desorption parameters are identified based on the surface isotherm test data. Based on NH_3 storage equilibrium calculations and transient adsorption data, the adsorption/desorption parameters and the total adsorption capacity of the catalyst are identified. The adsorption/desorption model is then validated using transient desorption data during TPD. During the second step, the model is calibrated for the remaining reactions using transient response test shown in Figure 1. Nonlinear minimization functions from Matlab's optimization toolbox are used to identify the parameters when transient data is used for calibration.

Results and Discussion

The catalyst model thus developed will be used for analysis and performance studies. Catalyst performance is defined as a function of NO_x conversion and NH_3 slip. Recent work on cu-zeolite[5] suggested that the optimal operating point for maximum NO_x conversion and minimal NH_3 slip in [NO/NO_2, NH_3/NO_x] is [1.0, 0.8] as shown in Figure 2.

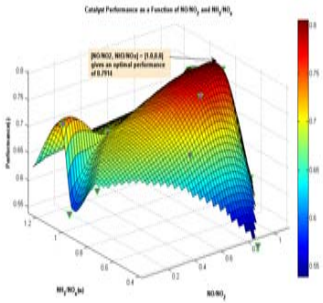

Figure 2: Catalyst Performance Studies on a Cu-zeolite catalyst

Significance

This work presents a two step approach to characterize an SCR catalyst using steady state data. Following the procedure, the catalyst can be calibrated, and the validated model can be used for analysis and controls design purposes.

References

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