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Oral presentation

Robust parameter estimation methodology for heterogeneous catalytic reactors

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Abstract
Modeling of Exhaust-Gas Aftertreatment Systems is an important tool for improved understanding and thus improved performance and durability. The challenges for accurate modeling of the multi-scale reactor are many and one important challenge is the interplay between mass transfer and kinetics. Although intrinsic kinetics (without effects from mass transfer) are possible to obtain by analysis of the washcoat separately, many challenges (e.g. washcoat distribution, ageing effects) are best studied using the monolith reactor structure.

In this study, a 1+1D diesel oxidation catalyst model was tuned to synthetic catalyst activity test (SCAT) bench data using a robust parameter estimation algorithm based on response surface methodology (RSM). The final residuals (SSE) were compared with experimental uncertainties to enable a statistical F-test to assess the model fit. Two different design of experiment (DoE) design matrices were compared to evaluate potential interaction effects between parameters. While the choice of DoE had different benefits, problems with each design could easily be circumvented.
Several parameter estimation cases were compared to investigate the importance of some key algorithm choices:

(a) the choice of a weight function for the residual calculation. A weight function sensitive to the experimental observation distribution obtained different fits with different parameter sets.

(b) The importance of carefully designed experimental observations. Simulations with catalysts containing an inert washcoat layer proved invaluable for tuning of internal mass transfer coefficients.

(c) The importance of experimentally measured constants as initial guesses. The use of intelligent gravimetric analysis (IGA) showed to give a much more suitable initial guess for tortuosity compared to literature data.

For all cases, the model fit gave insignificant F-test values (experimental uncertainties were larger than the model residuals), rendering that none of the parameter sets could be rejected. To demonstrate the significance of the different cases, the final parameter set for each case were compared through comparison of ratios of classical timescales, showing the experimental conditions for the various controlling regions of mass transfer and kinetics.