Motivation

- Monolith reactors are used in many catalytic after-treatment systems (e.g. TWC, DOC, LNT and SCR). These systems are often studied using monolith samples of the same cell density, washcoat thickness and catalyst loading but of a smaller length compared to the full scale systems. The space time is maintained the same in different scales in order to obtain similar performances.
- However, similarity may not exist and the ignition/extinction behavior of the two systems can be qualitatively different when thermal effects are significant (adimetric temperature rise is large) and heat and mass transfer and dispersion effects are not negligible.

Objectives

- Use modeling and dimensional analysis to quantify the impact of heat and mass dispersion and thermal effects on reactors with different scales.
- Develop criteria for similarity for experimental designs based on simulation and modeling results.

Model Equations

\[ \frac{\partial y_f}{\partial t} = \frac{1}{P_{m,f}} \frac{\partial^2 y_f}{\partial z^2} + \frac{1}{P_m} (y_f - y_{wc}) - \frac{\partial y_f}{\partial z} \]

\[ \frac{\partial y_{wc}}{\partial t} = \frac{1}{P_{m,wa}} \frac{\partial^2 y_{wc}}{\partial z^2} + \frac{1}{P_m} (y_{wc} - y_{wa}) - D_{wa} \frac{\partial y_{wa}}{\partial z} \exp \left(-\frac{1}{\beta} \frac{T_f}{T_{in} + \theta_m} \right) \]

Energy balance

\[ \frac{\partial T_f}{\partial t} = \frac{1}{P_{h,f}} \frac{\partial^2 T_f}{\partial z^2} + \frac{P_m}{P_{h,wa}} (T_f - T_{wa}) - \frac{\partial T_f}{\partial z} \]

\[ \frac{\partial T_{wa}}{\partial t} = \frac{1}{P_{h,wa}} \frac{\partial^2 T_{wa}}{\partial z^2} + \frac{1}{P_m} (T_f - T_{wa}) + D_{wa} \frac{\partial y_{wa}}{\partial z} \exp \left(-\frac{1}{\beta} \frac{T_f}{T_{in} + \theta_m} \right) \]

Dimension Analysis

\[ \tilde{t} = \frac{t}{L/\tilde{u}} \quad \frac{z}{\tilde{L}} \quad \tilde{y} = \frac{y}{\tilde{C} \tilde{m}} \quad \tilde{T} = \frac{T - T_{in}}{\Delta T_{ad}} \quad \tilde{g} = \frac{\theta_m}{\Delta T_{ad}} \]

Impact of Heat Conduction on Light-off

- Fitting data using inappropriate model can lead to false kinetic parameters.
- For example, using CSTR model to fit data from a reactor with high Pe_m (close to PFR) will result in \( E_a = 1.58 \Delta E_a \quad A = 1.718 \Delta A^{1.582} \)

Impact of Mass Dispersion on Light-off

- For linear kinetics, mass dispersion only cause quantitative differences, thus there can be similarity provided that the Pe_m are large enough in both scales.
- For Langmuir-Hinshelwood kinetics (negative order), mass dispersion can cause qualitative differences (multiple solutions).

Simulation Conditions

- Since reactors in different scales have the same catalyst loading and space time, a constant typical value of Da_0 = 10^10 and E_a/R = 12000 are used for all simulations. Similarly, P_m = P_h = 0 except for the hysteresis locus calculations, since they’re matched in different scales and are usually very small.
- \( \Delta T_{ad} = 200K \) is used for steady-state conversions (bifurcation diagrams), this can also be interpreted as 2% CO since \( \Delta T_{ad} = 10^\Delta T_{CO} \)

Summary

- Similarity between different scales generally doesn’t exist, thus a direct scale-up from lab scale is not possible.
- One should use the lab scale for kinetic studies only and make predictions of full scale system performances using modelling approach.
- For qualitative similarity, experiments should be designed such that lab scale reactors fall in the same multiplicity region as the full scale system.
- Mass and heat dispersion should be considered while estimating kinetic parameters to avoid false predictions.

Current Work

- In this work we only considered isothermal and adiabatic reactors, where the external heat transfer coefficient are considered to be either infinity or zero.
- Since one can not match the external heat transfer conditions in different scales, this can lead to more scale-up issues to consider.