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Development of a machine learning model to account for diffusional limitations in the simulation of highly exothermic reactions in an industrial reactor

Aim

The primary aim of this thesis is to develop a machine-learning surrogate model that approximates a detailed submodel describing diffusion limited reaction kinetics in catalyst pellets. The model will be integrated into a transient 2D pseudo-homogeneous reactor model designed to simulate highly exothermic reactions in multi-tubular reactors, thereby enabling the prediction of catalyst deactivation.

Justification

Multi-tubular reactors are essential for managing highly exothermic reactions in a variety of industrial processes, including ethylene oxidation, phosgene synthesis, and CO_2 methanation, primarily due to their efficient heat transfer capabilities. In these reactions, the rapid generation of heat leads to the formation of hot spots, which in turn can cause catalyst deactivation. As a result, the catalyst has to be replaced at regular time intervals, which has a negative impact on the operational time of the reactor. To achieve a more efficient reactor operation, simulation models can be recurred to, which require an accurate description of the reaction kinetics (including deactivation) as well as the heat and mass transport phenomena.

The current pseudo-homogeneous reactor model assumes uniform conditions within the catalyst pellet. This simplification neglects the internal diffusional resistance, leading to an overestimation of the overall conversion. The heat transfer parameters will compensate for this during regression to reproduce the experimentally observed temperature profile. However, this results in inaccurate predictions when operating conditions, such as the cooling temperature, are modified.

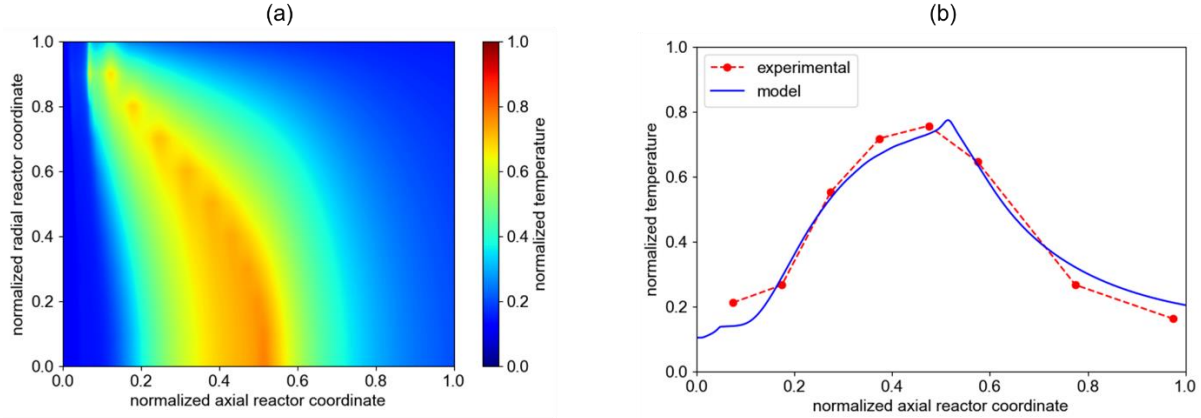

Figure 1: (a) Two-dimensional and (b) centerline temperature profiles after 60 days of operation, as predicted by the current pseudo-homogeneous reactor model, which neglects diffusional limitations within the catalyst pellets.

It is therefore crucial to account for internal diffusional limitations and the impact on the overall conversion. A detailed submodel that resolves these gradients could be added, but its computational complexity makes it impractical for integration into full-scale, transient reactor simulations. As an alternative, a machine learning surrogate model can be trained to emulate the detailed submodel, offering a rapid yet accurate alternative that is computationally efficient enough to be embedded within a transient 2D reactor simulation framework.

Program

1. Literature survey of modeling techniques for heterogeneous catalytic reactions in multi-tubular reactors, with a focus on diffusional limitations within catalyst pellets. This will include examining machine learning methods used to develop surrogate models.
2. Development of a detailed model in Python for the catalyst pellet that accounts for internal diffusional resistance. Subsequently, a machine learning surrogate model will be developed to approximate the detailed model, by training it on data generated by the detailed model simulations.
3. Incorporation of the surrogate model into an existing transient 2D pseudo-homogeneous reactor model framework. The integration aims to improve the prediction of reactor behaviour particularly catalyst deactivation and temperature profiles under various operating conditions. The resulting reactor model can be validated against experimental data, such as the observed temperature profiles after extended operation.