Development of a surrogate model for microkinetic modeling of the CO_x methanation reaction and fine-tuning using transfer learning

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Introduction

Optimization of catalytic reactors requires a suitable description of the reaction rate. Simple power-law expressions offer the simplest representation of heterogeneously catalyzed systems. However, power-law expressions allow only for a limited fundamental description of the catalyst surface processes, which restricts their range of validity. In contrast, microkinetic elementary step models, such as the one developed by Schmider et al. [1] for CO_x methanation, represent the system as a detailed network of surface interactions. The accuracy achieved with these models comes at the expense of high computational load. A traditional technique for the reduction of a microkinetic mechanism is the use of Langmuir-Hinshelwood-Hougen-Watson kinetics, which focus on the elementary steps that most significantly affect global kinetics. In recent years, deep learning has gained attention as an alternative to these traditional kinetic modeling methods. Deep learning models can serve as surrogates to simulate the behavior of a microkinetic system, increasing the speed of calculations [2].

While studies of heterogeneous catalysis and the models developed often align with experimental data, their accuracy tends to deteriorate outside the conditions under which they were obtained. As a potential solution to this issue, transfer learning (TL) is explored in this work. This methodology, which involves adapting pre-trained deep networks to smaller training data sets, has the potential to significantly reduce the modeling and experimental effort required to fit new models to specific systems.

Methods

Based on the Global Reaction Neural Network (GRNN) architecture by Kircher et al. [2] a surrogate model for the microkinetic elementary step model by Schmider et al. [1] is developed. The resulting model is adapted to the experimental data of Langer et al. [3], who used a catalyst different than that considered during the development of the microkinetic model.

Results

We were able to successfully approximate the steady-state reaction rates of the microkinetic model using a neural network (NN). As can be seen in Figure 1, after adjusting the active specific surface area (SSA) to the experimental data from Langer et al. [3], the NN without TL already approximates the experimental data well, but an adaptation is desirable. To show that only a part of the experimental data is needed during TL, we use experimental data up to a temperature of 350 °C or 400 °C. It can be seen that the model is in very good agreement with the experimental data used for training and also extrapolates very well.

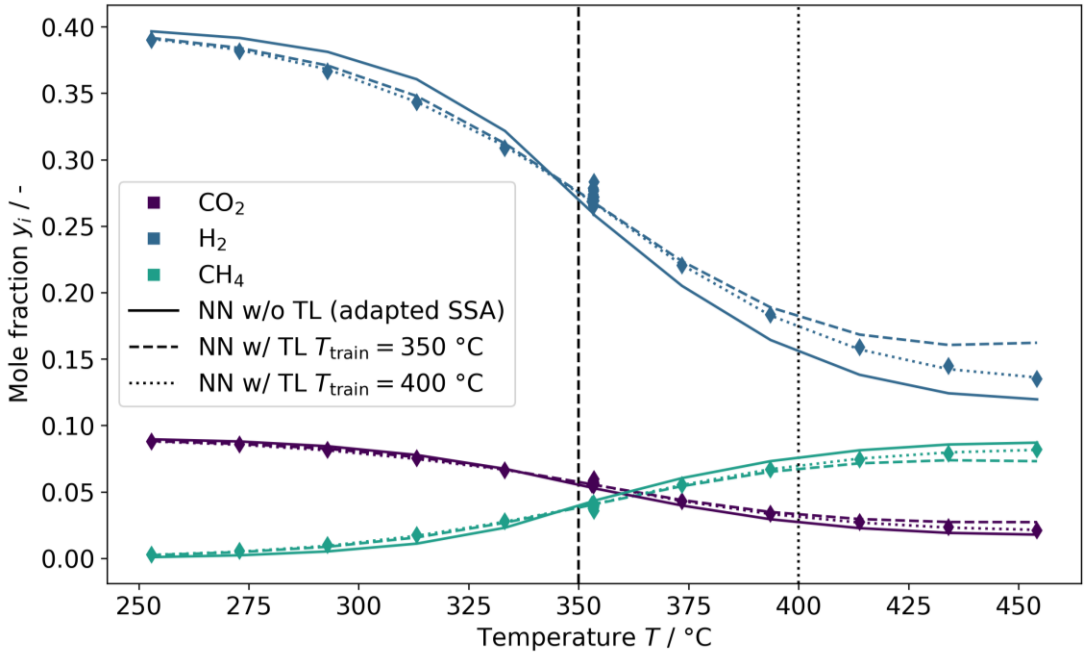

Figure 1: Steady-state trajectories of y_i ($i \in \{CO_2, H_2, CH_4\}$) over the temperature at 10 bar, 8 ml_{STP} min^{-1} mg^{-1}_{cat} with a feed containing H_2:CO_2:N_2 in a ratio of 4.4:1:5.6 in a CSTR. Markers indicate experimental data from Langer et al. [3]. The solid line represents the prediction using the surrogate model without transfer learning and the dashed and dotted lines represent the prediction by the adapted surrogate models with training data up to T_{train} = 350 °C and T_{train} = 400 °C, respectively.

References

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