Accelerating Kinetic Model Discovery by Global Reaction Neural Networks with Embedded Stoichiometry and Equilibrium

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Highlights

- Global Reaction Neural Networks learn kinetics from noisy integral reactor data
- Due to the embedded physics, they outperform conventional neural networks
- Successful application to both systems with equilibrium limitations and stiff kinetics

1. Introduction

The digitalization in chemical research, alongside big data frameworks [1], has led to an abundance of data for developing kinetic models. But model parameterization is not keeping up with the data supply and is becoming the bottleneck in the model-based engineering workflow. Machine learning tools are urgently required to exploit the potential of this data. Neural ordinary differential equations (neural ODEs) [2] recently emerged as the state-of-the art for learning dynamics from time-series data, such as integral reactor measurements, by neural networks. We introduce Global Reaction Neural Networks (GRNN) [3] with embedded physico-chemical knowledge of stoichiometry and chemical equilibrium and train them as a neural ODEs on integral reactor data. We show that the models accurately retrieve the true kinetics from noisy integral reactor measurements, both for equilibrium limited reactors [3] and for reactors with stiff kinetics operating close to or at full conversion [4].

2. Methods

Global Reaction Neural Networks with embedded stoichiometry and thermodynamics map reaction conditions to a latent representation of global reactions rates, which is then used to calculate the corresponding chemical source terms by embedded algebraic equations [3]. When coupled with reactor physics, they can be trained as neural ODEs on integral reactor data. The neural network layers of the GRNN map the partial pressure p and temperature T to latent representations of the forward reaction rates $\overrightarrow{r}$. The De Donder relation is then used to compute thermodynamically consistent net reaction rates r^{net} from the forward rates using tabulated thermochemistry. These net rates approach zero if reactants are depleted and predict the correct equilibrium. Then, the chemical source terms $\dot{s}$ are calculated using the embedded stoichiometry. These source terms strictly conserve the atom balance. The model architecture, including the embedding algebraic equations, is shown in figure 1.

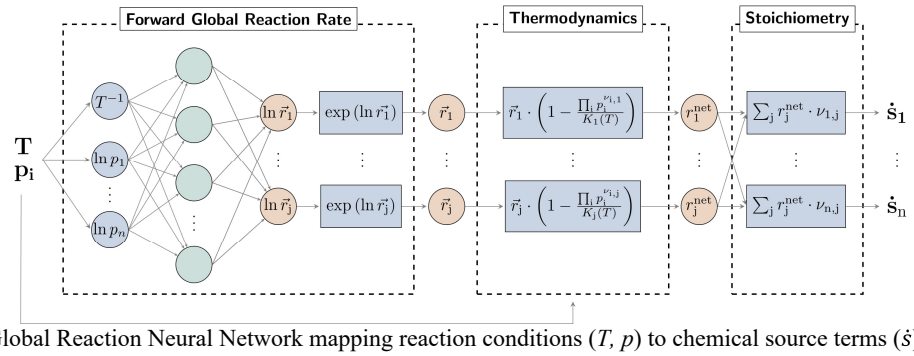

Figure 1. Global Reaction Neural Network mapping reaction conditions (*T, p*) to chemical source terms ($\dot{s}$). Embedded physical knowledge is blue, latent variables of interest are red and latent variables of the hidden layers are green [3].

We showcase our modelling approach for two different reaction systems. We use a steam reforming reactor for anode-off gas recycling of a fuel cell, operating close to the equilibrium, and a reactor for the preferential oxidation of CO for pretreatment of streams entering a fuel cell, operating close to or at full conversion, as representative application examples. For both we defined a range of input conditions

where we required our machine learned kinetic model to work and randomly sampled inlet conditions from this range. Synthetic experimental data of the reaction systems was generated for these conditions with 1-D plug flow reactor models, using the microkinetic model by Maestri et al. [5] and Hauptmann et al. [6]. A laboratory setting was mimicked by reducing synthetic experimental data to mole flows and temperature measurements at six equidistant points along the reactor length and adding gaussian noise.

3. Results and discussion

We used our proposed method for automated kinetic modelling to separately learn kinetics both from an equilibrium limited system [3] (Steam Reforming) and a reactor operating close to full conversion (Preferential oxidation of CO) [4]. In the first step, we demonstrated that the neural ODE with embedded physics accurately retrieved the kinetics from reactor experiments without noise, whereas conventional neural ODEs failed. In the second step, we trained our global reaction neural ODE on reactor experiments with significant noise. Here, we showed that for both reaction systems the models recovered the true solution with higher quality than the noisy data used for training it and generalized to unseen data. This is because the models are highly biased towards the true solution due to the embedded physics. Figure 2 shows an example from the preferential oxidation of CO with ground truth (dots), noisy training data (diamonds), and predictions of the trained GRNN (lines) for the O_2 and CO molar flows (left) and the O_2 source terms (right). The global reactions governing the system are shown on the right. The model was only trained on the noisy data and did not have access to ground truth molar flows or source terms.

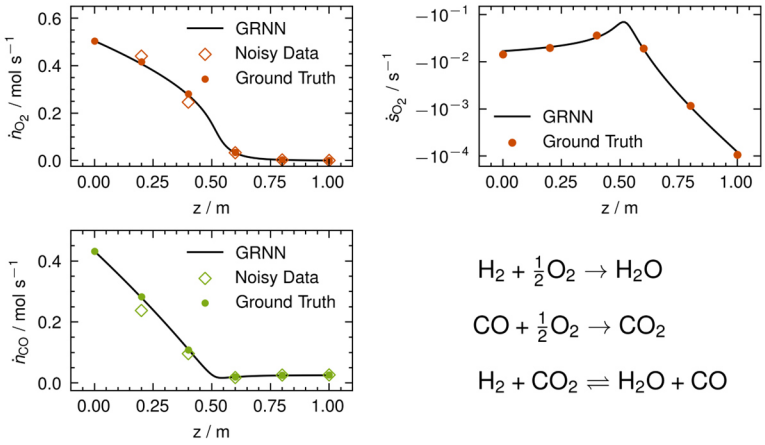

Figure 2. Profile of O_2 and CO molar flow (left) and O_2 source terms (right) predicted by the trained GRNN (line), the noisy training data (diamonds) and the ground truth (circles) [4].

4. Conclusions

Data availability is increasing due to digitalization, high-throughput experimentation, and new data infrastructures. Our proposed approach allows the combination of this data with prior knowledge for the autonomous generation of kinetic models. These kinetic models generalize well to unseen data and accurately recover the ground truth chemical source terms. As our approach has proven successful for two important edge cases of industrial applications, namely equilibrium limitation and high conversion, we anticipate that it will be applicable to a wide range of systems and thus contribute to the acceleration of kinetic model development.

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Keywords
Kinetic Modelling, Digitalization, Machine Learning, Neural ODE