Optimization of the Syngas Combustion Chemistry Model

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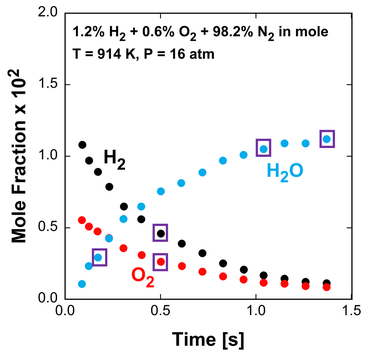
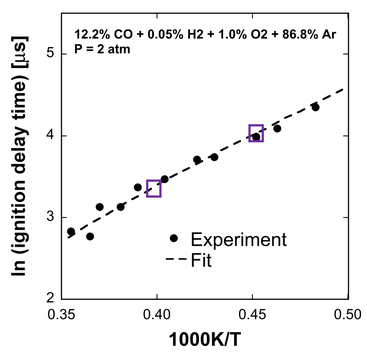
Motivation

- Synthetic gas (syngas) consists of hydrogen (H_2) and carbon monoxide (CO). An accurate syngas combustion chemistry model is of great importance when it is combined with the computational fluid dynamics (CFD) simulations to predict engine performance.
- A typical syngas combustion chemistry model contains about 30 reactions (see solution mapping section). In each reaction, the rate parameters were either determined by direct measurements or quantum chemistry calculations. Since the parameters were obtained independently, neglecting the couplings between each other, the model might not achieve great accuracy.
- In the current work, we present an optimization approach to improving the performance of the syngas combustion chemistry model. To realize this goal, we use earlier global combustion experimental data as targets to optimize the rate parameters of the syngas model. Result shows that the optimized model can achieve better prediction accuracy than the original model.

Experimental Data

- Typical combustion properties include species concentration profiles, ignition delay time, laminar flame speed, etc..
- Two selected data sources are shown below [1, 2]. The training and test data are extracted from the full dataset (marked as open squares in the following figures). The selected data should be enough to represent the characteristics of the original experimental data curves. In this study, we have in total of 200 experimental data points.





Solution Mapping – Response Surface Method

- The syngas model (shown below) is first built without optimization. The rate parameters (A, n, E_a) are obtained from [3].

No.	Reactions	Reaction rate coefficients A (mol/cm³,s)	n	E_a (cal/mol)	Uncertainty factor, f
1	H+O2 = O+OH	2.65E+16	-0.671	17041.0	1.15
2	O+H2 = H+OH	3.87E+04	2.700	6260.0	1.3
3	OH+H2 = H+H2O	2.16E+08	1.510	3430.0	1.3
4	OH+OH = O+H2O	3.57E+04	2.400	-2110.0	1.3
5	H+H+M = H2+M	1.00E+18	-1.000	0.0	2
6	H+H+H2 = H2+H2	2.20E+22	-2.000	0.0	2
7	H+H+H2O = H2+H2O	4.71E+18	-1.000	0.0	2
8	H+H+CO2 = H2+CO2	1.20E+17	-1.000	0.0	2
9	H+OH+M = H2O+M	4.65E+12	0.440	0.0	1.2
10	O+H+M = OH+M	7.40E+05	2.433	53502.0	1.25
11	O+O+M = O2+M	7.40E+13	-0.370	0.0	1.5
12	H+O2(+M) = HO2(+M)	3.97E+12	0.000	671.0	1.5
13	H2+O2 = HO2+H	7.08E+13	0.000	295.0	2
14	OH+OH(+M) = H2O2(+M)	2.00E+13	0.000	0.0	2
15	HO2+H = O+H2O	2.90E+13	0.000	-500.0	2
16	HO2+H = OH+OH	1.30E+11	0.000	-1630.0	1.5
17	HO2+O = OH+O2	1.21E+07	2.000	5200.0	2
18	HO2+HO2 = O2+H2O2	2.41E+13	0.000	3970.0	2
19	OH+HO2=H2O+O2	9.63E+06	2.000	3970.0	2
20	H2O2+H = HO2+H2	2.00E+12	0.000	427.0	2
21	H2O2+H = OH+H2O	1.80E+10	0.000	2384.0	2
22	H2O2+O = OH+HO2	7.05E+04	2.053	-355.7	2
23	H2O2+OH = HO2+H2O	2.53E+12	0.000	47700.0	3
24	CO+O(+M) = CO2(+M)	3.01E+13	0.000	23000.0	2
25	CO+OH = CO2+H	1.20E+14	0.000	0.0	2
26	CO+O2 = CO2+O	3.00E+13	0.000	0.0	2
27	CO+HO2 = CO2+OH	3.00E+13	0.000	0.0	2
28	HCO+H = CO+H2	3.02E+13	0.000	0.0	2
29	HCO+O = CO+OH	9.35E+16	-1.000	17000.0	2
30	HCO+O = CO2+H	1.20E+10	0.807	-727.0	2

The reactions in red are effective reactions obtained through sensitivity analysis in a Fortran simulator called ChemKin [4]. Those effective reactions are treated as the features in the current study.

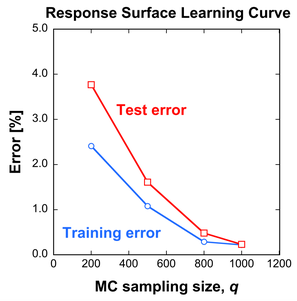
Traditionally, the chemistry model is implemented with ChemKin to calculate combustion properties. In the current study, we use the response surface method bypassing the ChemKin simulator.

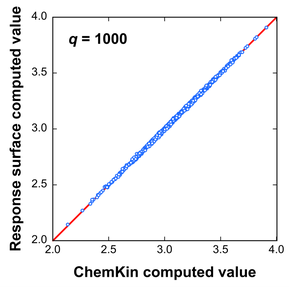
- We randomly perturb the features q times under each experimental condition, and compute the model response $\eta_{resp}^{(i)}$ using each random model through ChemKin. Then we represent $\eta_{resp}^{(i)}$ by a second-order polynomial:

$$\eta_{resp}^{(i)} = c^{(i)} + \sum_{j=1}^{n} b_j^{(i)} x_j^{(i)} + \sum_{j=1}^{n} \sum_{k \geq j}^{n} a_{jk}^{(i)} x_j^{(i)} x_k^{(i)}$$

- x is the feature variable. The coefficients, $c^{(i)}$, $b_j^{(i)}$, and $a_{jk}^{(i)}$ are obtained by directly solving the normal equations.
- Test shows 1000 Monte Carlo sampling points are enough for generating accurate response responses.

Monte Carlo sampling size, q	q = 200	q = 500	q = 800	q = 1000
Training error (%)	2.41	1.08	0.29	0.22
Test error (%)	3.77	1.61	0.48	0.23





Model Optimization

Optimization Approach 1: Locally weighted least square

- The optimization step can be carried out through:

$$\text{minimize} \quad J(x) = \sum_{i=1}^{m} \left[\frac{\left(\eta_{expt}^{(i)} - \eta_{resp}^{(i)}\right)}{\sigma_{expt}^{(i)}} \right]^2$$

- $\eta_{resp}^{(i)}$ is the model response, $\eta_{expt}^{(i)}$ is the experimental data point, and $\sigma_{expt}^{(i)}$ is the experimental uncertainty factor of the i^{th} data point.
- The optimization are carried out using gradient descent method.

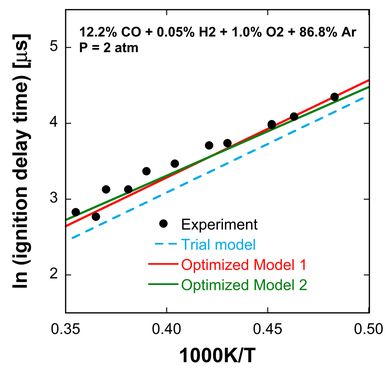
Optimization Approach 2: Reinforcement learning

- For each data point, model has three outcomes: overprediction, underprediction, or falling within its uncertainty band. For m data points, $|S| = 3^m$. The actions are to increase/decrease the parameters by 20% of its uncertainty factor, f. For n effective reactions, $|A| = 2^n$.
- If all the model predictions fall within data uncertainties, a positive reward is assigned; otherwise negative reward is assigned.
- The value function is updated using value iteration based on Bellman equation:

$$V^{\pi}(s) = R(s) + \gamma \sum_{s' \in S} P_{s\pi(s)}(s')\, V^{\pi}(s')$$

Optimized Model Performance

- One selected model validation case is shown on the right side. Note that model obtained through both approaches can better predict the experimental data.
- To save storage, the RL approach only used 5 data points instead of 200. In the future, we can extend the training to full data set.



References

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[4]. Kee, Robert J., Fran M. Rupley, and James A. Miller. Chemkin-II: A Fortran chemical kinetics package for the analysis of gas-phase chemical kinetics. No. SAND-89-8009. Sandia National Labs., Livermore, CA (USA), 1989.