A Physics-Constrained Neural Network Model for Combustion Chemical Kinetic Prediction

Tinghao Wang[1], Tianhan Zhang[2]*, Zheng Chen[1]
[1]SKLTCS, Department of Mechanics and Engineering Science, College of Engineering, Peking University
Beijing, China
[2]Department of Mechanics and Aerospace Engineering, SUSTech
Shenzhen, China

1 Introduction

Numerical simulation provides details in combustion processes and helps to reveal the physical-chemical mechanisms involved therein. Therefore, numerical simulation becomes one of the most important research tools for developing alternative fuels and high-performance combustion engines. To achieve quantitative rather than qualitative prediction, detailed kinetic model needs to be considered in simulations. However, the broad range of temporal scales and large number of species involved in kinetic model bring great challenge for combustion simulation. The computational cost increases greatly with the size of the kinetic model. Therefore, there is an urgent need to develop a high-efficient surrogate model for chemical kinetic modeling to improve the computation efficiency.

Artificial neural network has been widely known for its extraordinary ability to fit nonlinear, high-dimensional data [1]. It has been applied successfully in the combustion area since the 1990s [2]. The trained neural networks predict the transient evolutions of the chemical source term such that direct integration can be replaced efficiently. However, reaction systems with both low-temperature and high-temperature chemistry (LTC and HTC) bring new challenges to neural networks [3][4]. In a two-stage ignition process, the species concentration distribution has scale separation, and the characteristic timescales are divergent. In addition, the low-temperature ignition and cool flame rely on different radical chain branching pathways from the high-temperature chemistry cases, which is difficult for a traditional neural network to predict. Furthermore, only a few studies consider element conservation for the neural network-based model. The violation of the fundamental conservation law makes the method inaccurate and unstable.

The objective of this work is to construct a robust neural network model which keeps the conservation law and can predict LTC and HTC by a single neural network. The isobaric homogeneous autoignition problem is considered here. Dimethyl ether (DME)/air mixture is chosen since it has two-stage ignition and the LTC and HTC of DME are both well-studied. A kinetic model DME consisting of 39 species and 175 elementary reactions [5] is used. Based on the method developed by Zhang et al. [4], the Box-Cox Transformation is introduced to mitigate multi-scale effects in combustion. Data samples are

Correspondence to: zhangth@sustech.edu.cn

obtained by the manifold sampling method with a wide range of initial conditions, and a single neural network is trained offline. The accuracy of the neural network method is verified, and the computational efficiency of the neural network is explored.

2 Data Sampling and Neural Network Methods

The neural network aims to predict the average changing rate for each physical state at a large time step. The physical state of the gas mixture includes temperature T, pressure P and species mass fraction Y_i, which is a 41-dimensional vector denoted by X. The reaction system has the following form:

$$\frac{dX}{dt} = g(X) \tag{1}$$

In the study, we set a specific time step Δt. The neural network's task is to predict the time integration with a fixed time-step Δt. The procedure consists of the following four steps:

Step I, Sampling. Under a specific time step Δt, samples in the form of (X_0, X_1) are obtained by direct numerical simulation (Cantera solver [6]), where X_0 is the physical state at a certain moment, and X_1 is the physical state evolved by X_0 after a fixed time step Δt. The training data is assembled by simulating auto-ignitions at different initial conditions. In addition, the data is screened. Therefore, there are abundant samples with high heat release rates ($HRRs$).

$$p(selected) = \begin{cases} p_1 & HRR < HRR_0 \\ p_2 & HRR \geq HRR_0 \end{cases} \tag{2}$$

In Eq. (2) , p_1 and p_2 are probabilities of selecting data whose $HRRs$ are below or above HRR_0, a threshold to distinguish low and high heat release rates.

Step II, Data preprocessing. For the mass fraction of species Y_i, Box-Cox Transformation (BCT) [7] is adopted. This transformation can mitigate the multi-scale effect of combustion data. The formula of BCT is [7]:

$$f(x) = \begin{cases} log\, x\,, \lambda = 0 \\ \frac{x^{\lambda}-1}{\lambda}\,, \ \lambda \neq 0 \end{cases} \tag{3}$$

For all species, we set the parameter $\lambda = 0.1$. Denote $\widetilde{X_0}$ and $\widetilde{X_1}$ as the data transformed with BCT and with standard normalization. The neural network's output is $\widetilde{X_1} - \widetilde{X_0}$.

Step III, Neural network training. The neural network has four hidden layers, and the numbers of neurons are 800, 1600, 800, and 400. The activation function is GELU [8], and the loss function is L_1.

Step IV, Inference. By using the transformation in step II and the neural network trained in step III, the neural network predicts the value of X_1 from X_0. The predicted values of 38 species except N_2 are used, and the mass fraction of N_2 is calculated from other species, the pressure P equals the initial pressure (isobaric problem), and the temperature T is calculated based on energy conservation.

3 Results and Discussions

3.1 The Result of Manifold Sampling

The range of initial conditions selected in this study is as follows: initial temperature $T = 700 - 1200\ K$, initial pressure $P = 5 - 20\ atm$, equivalence ratio $\varphi = 0.8 - 1.2$. 8000 initial conditions are randomly generated, and Cantera is used for DNS and data sampling. A specific time step $\Delta t = 10^{-6}\ s$ is selected. The parameters of the sampling probability function are $p_1 = 0.01$, $p_2 = 0.3$ and $HRR_0 = 10^8\ J/(kg \cdot s)$. About two million data points are selected with 10% for the test set and the other 90% for the training

set.

Figure 1 is the phase diagram for the sampled data. The x-axis is the magnitude of each physical quantity, and the y-axis is the average time change rate of the physical quantity in a single time step Δt. The phase diagram effectively shows the distribution characteristics of sampled data. The physical quantities displayed include temperature and mass fractions of 8 species. The points are colored by temperature. The multi-scale effect of evolution of physical quantities brings challenges to neural network training.

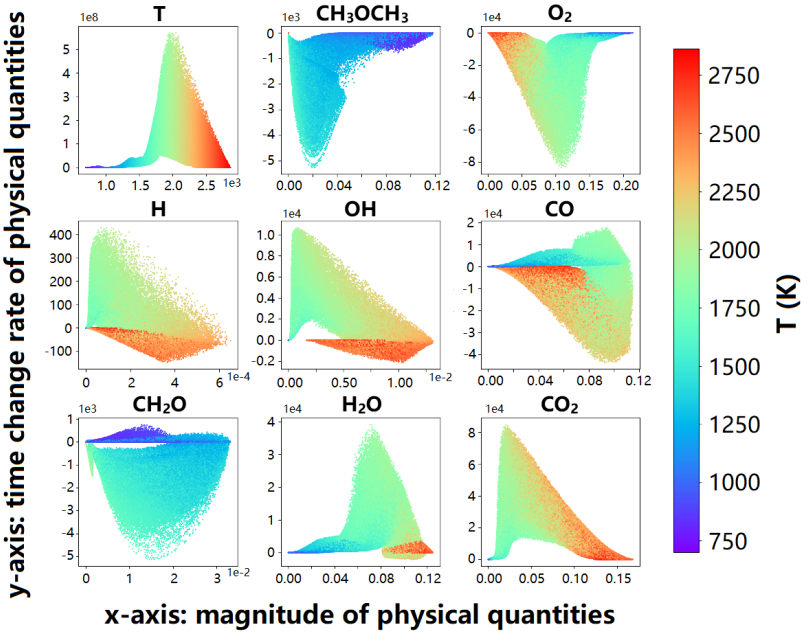

Figure 1: Phase diagram of sampled data, where the x-axis represents the magnitude of each physical quantity, and the y-axis represents its time change rate. The scatter plot is colored by temperature.

3.2 Prediction Accuracy of the Neural Network

Figure 2 shows the predicted evolutions of temperature, mass fraction of DME, and mass fraction of HO_2 under the initial condition of $T = 750\,K$, $P = 10\,atm$ and $\varphi = 1.0$. The neural network's prediction (green) agrees well with the DNS simulation based on the detailed chemical mechanism (yellow). Near the igniting moment when chemical species react violently, the neural network accurately predicts the temporal evolution of each physical quantity with a time step much larger than the built-in time step of the Cantera solver (on the order of 10^{-8} s).

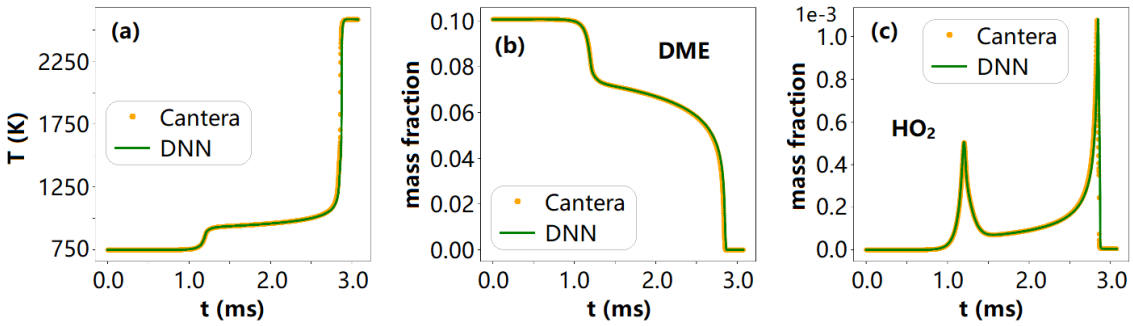

Figure 2: Temporal evolution of (a) temperature T, (b) mass fraction of DME and (c) mass fraction of HO_2 for the initial condition of $T = 750\,K$, $P = 10\,atm$ and $\varphi = 1.0$.

Figure 3 shows the predictions of autoignition delays at different initial temperatures with $P = 10\ atm$ and $\varphi = 1.0$. The neural network successfully predicts autoignition delays, including low-temperature ignition (LTI) and high-temperature ignition (HTI). Compared to results predicted by Cantera, the ignition delays predicted by the neural network have a mean relative error of 0.4% and a maximum relative error of 2.6%. Therefore, the two-stage ignition is accurately predicted by the neural network.

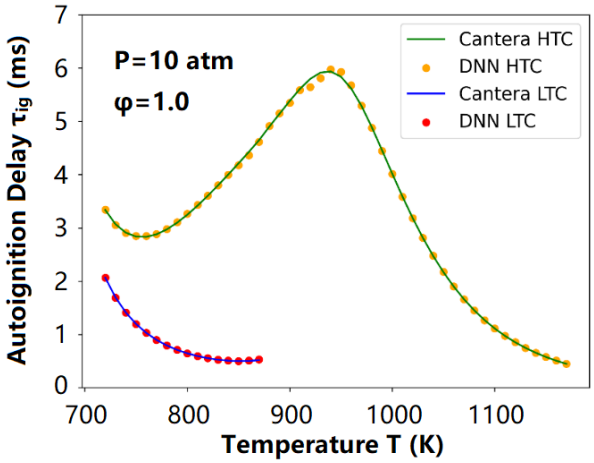

Figure 3: Predictions of autoignition delays at different temperature T for fixed $P = 10\ atm$ and $\varphi = 1.0$.

In conclusion, a single neural network can learn the characteristics of LTC and HTC. The well-trained DNN can accurately predict the temporal evolution of physical quantities within a wide range of initial conditions.

3.3 Physical Conservation

In the above method, the neural network does not guarantee the conservation of elements. As the neural network method advances multiple time steps, the errors of elements accumulate and may lead to unstable prediction. A new neural network is trained based on the initial temperature $T = 1000 - 1200\ K$, initial pressure $P = 0.5 - 2\ atm$, and equivalence ratio $\varphi = 0.8 - 1.2$. The time step remains to be $\Delta t = 10^{-6}\ s$.

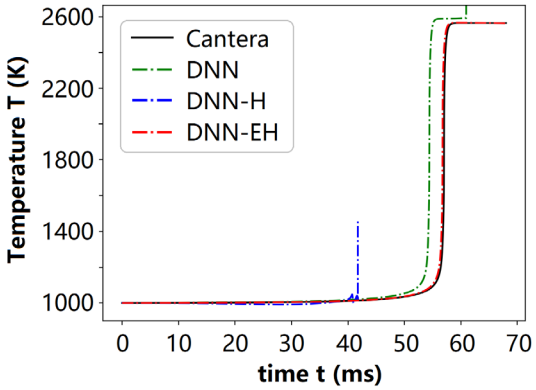

Figure 4: A test case with initial condition $T = 1000\ K$, $P = 0.7\ atm$ and $\varphi = 1.0$.

Figure 4 shows a test case at $T = 1000\ K$, $P = 0.7\ atm$ and $\varphi = 1.0$. Without considering element conservation, the neural network goes divergent (blue line). However, if the neural network predicts the temperature instead of calculating based on energy conservation, the neural network performs better (green line). The temperature T predicted by energy conservation leads to an additional error of temperature T.

We apply a correction method to ensure the conservation of elements. The final prediction of the neural network method, denoted as X_1^{DNN}, leads to errors of elements e_i

$$e_i = \sum_j a_{i,j}(X_1^{DNN} - X_0)_j \tag{4}$$

In Eq. (4), i represents all the elements, and j represents all the species. $a_{i,j}$ is the mass fraction of the i-th element in the j-th species. The T and P components of X are neglected because they have no relationship to element conservation. We choose the correction term

$$\Delta X_0^{cor} = \mathrm{argmin}_{\sum_j a_{i,j}(\Delta X)_j + e_i = 0} \sum_j [(X_0)_j^{\lambda-1}(\Delta X)_j]^2 \tag{5}$$

The right-hand side of Eq. (5) is a least square problem for ΔX and can be solved analytically. The parameter λ is just the one applied in BCT. Setting $X_1 = X_1^{DNN} + \Delta X_0^{cor}$ as our new prediction, the conservation of elements is ensured. When both element conservation correction and energy conservation correction are applied, the neural network method works perfectly during the whole case (red line). The conservation of elements promotes the robustness of the neural network method.

3.4 Computational Efficiency

Cantera and the neural network are used to predict randomly selected data points for a single time step, and the average CPU time is calculated. Unlike the CVODE solver, the neural network is based on matrix multiplication and addition, which naturally has the advantage of parallel computing (predicting the evolution of multiple data simultaneously).

Figure 5 shows the computational efficiency. It is seen that the computational efficiency of the neural network is about four times that of Cantera in terms of the efficiency of serial computing (predicting one by one). Furthermore, the computational efficiency of the neural network can be increased by more than an order of magnitude through parallel computing. The multi-point parallel computing may play an important role in the high-dimensional numerical simulation to improve the efficiency of the neural network method.

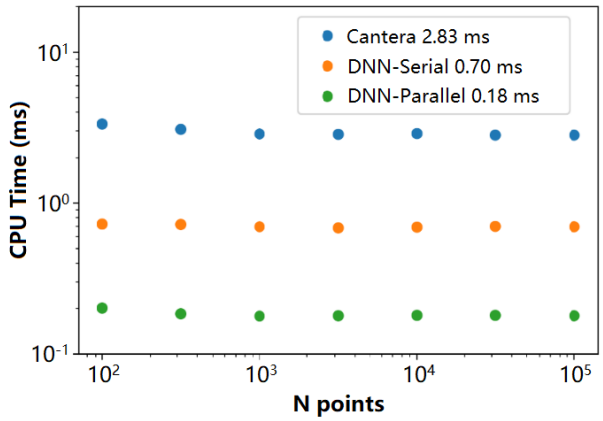

Figure 5: Computational efficiency of Cantera and neural network.

4 Conclusions

This study introduces the neural network method to solve the chemical source term. The method is demonstrated by solving isobaric homogeneous ignition in a DME/air mixture. The main conclusions are as follows:

(1) The neural network method combined with BCT overcomes the multi-scale effect of data. It can accurately predict the time change rate of each physical quantity in thousands of steps of temporal evolution, using a time step far larger than the time step used by a traditional ordinary differential equation solver.

(2) In a wide range of initial conditions, the neural network can accurately predict both two-stage and one-stage autoignition. LTC and HTC of DME can be accurately predicted by a single neural network.

(3) The current neural network guarantees the conservations of mass, energy, and element, which is a fundamental requirement for the any surrogate model but frequently ignored by previous machine learning-related modeling. The physics-constrained network improves the robustness and training efficiency of the neural network method.

(4) Compared with the Cantera solver, the computational efficiency of the neural network method increases by one order of magnitude. If parallel computing is combined for efficient calculation, the neural network is expected to improve the computational efficiency to a greater extent for complex combustion simulations.

This study demonstrates the promosing application of neural network in improving the efficiency of chemical calculation. Combining the physics constraint to design a better neural network structure to improve robustness and stability is an interesting yet challenging task that needs more investigation in the future works.

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