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MODELING JONES' REDUCED CHEMICAL MECHANISM OF METHANE COMBUSTION WITH ARTIFICIAL NEURAL NETWORK

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ABSTRACT

The present work reports a way of using Artificial Neural Networks for modeling and integrating the governing chemical kinetics differential equations of Jones' reduced chemical mechanism for methane combustion. The chemical mechanism is applicable to both diffusion and premixed laminar flames. A feed-forward multi-layer neural network is incorporated as neural network architecture. In order to find sets of input-output data, for adapting the neural network's synaptic weights in the training phase, a thermochemical analysis is embedded to find the chemical species mole fractions. An analysis of computational performance along with a comparison between the neural network approach and other conventional methods, used to represent the chemistry, are presented and the ability of neural networks for representing a non-linear chemical system is illustrated.

INTRODUCTION

Direct integration of the governing differential equations of a chemical mechanism with various reactive species requires too much computer processing time and also the complexity of chemical kinetics and interactions between chemistry and governing combustion equations are one of the outstanding difficulties for handling the engineering applications in this field. With the exception of direct integration method, which needs extensive computing time, several methods have been introduced to handle chemical systems, such as Look-up tables [1] and In-situ adaptive tabulation [2]. These techniques have various numerical costs. For example, Look-up tables store the incremental changes in composition, due to the reaction. So, this approach has the disadvantage of large memory requirements.

A neural network is a parallel dynamic structure and neural modeling of complex systems is an expanding field of application and research. The neural modeling is to adjust the synaptic weights of the network during an iterative process in the training phase. Then the neural network learns the principles of the physical problem and generates the required model. This model provides an approximation of the actual system with the same physical and mathematical behaviors.

With the exception of neural networks abilities of parallel computing and learning, they have also two main advantages: first, neural networks are fault tolerant against noises, which are appearing in many engineering applications, and second, since neural network is made of large number of interconnected neurons, the network is not dependent to a particular neuron and the presumptive errors of neurons will be damped during the parallel computing process.

In this work, the temporal changes of reactive chemical species, in Jones and Lindstedt chemical mechanisms, have been modeled by a feed-forward neural network [3]. Although, the application of neural networks to chemical mechanisms has been reported in recent works (Blasco et al. [4] and Christo et al. [5]), the main motivations for doing such a work were to model different reduced chemical mechanisms for methane combustion and making a comparison between the results from the point of view of CPU time and RAM memory for different mechanisms in the future. Blasco et al. [3] used the neural network method to model the temporal changes of seven

reactive species for another important reduced chemical mechanism, which was introduced by Peters and Kee [6]. Peters and Kee chemical mechanism is methane/air combustion mechanism with nitrogen, as an inert non-reactive species, but in the present research, we have neglected the effects of nitrogen, as an inert species, and assumed a methane/oxygen combustion mechanism with the stoichiometric mixture fraction of $f_{st.} = 0.2$.

In the last section of this work, an analysis of computational costs and performance and a comparison between different approaches are presented.

NOMENCLATURE

E_t	total error of the neural network
f	mixture fraction
$f()$	activation function
n	moles per mass of mixture
y	expected output of the network
$\hat{y}$	estimated output of the network
α	momentum factor
η	learning rate
θ	bias value
ω	synaptic weight

The Chemical Mechanism

The reduced chemical reaction, which is considered in the present neural modeling, is Jones and Lindstedt reduced chemical reactions for methane combustion. As we know, for extracting reduced chemical mechanism from detailed chemistry, some of minor elementary reactions and intermediate species are eliminated and also some species are assumed to be in partial equilibrium state.

This four-step mechanism was built by extracting six reactive species and simplifying them into four-step reactions. Jones and Lindstedt reported that this four-step mechanism is an acceptable mechanism and it is applicable to both diffusion and premixed laminar flames. Moreover they confirmed that the obtained burning velocity and flame structure (such as temperature and species concentration distributions) inside the flame conform well to actual measurements.

The original four-step reaction mechanism was proposed for generic saturated lower-hydrocarbon fuels, and for the case of methane, the mechanism is proposed as follows:

$$\begin{aligned} &R1: \quad CH_4 + H_2O \rightarrow CO + 3H_2 \\ &R2: \quad CH_4 + \frac{1}{2}O_2 \rightarrow CO + 2H_2 \\ &R3: \quad H_2 + \frac{1}{2}O_2 \leftrightarrow H_2O \\ &R4: \quad CO + H_2O \leftrightarrow CO_2 + H_2 \end{aligned} \tag{1}$$

The reaction rate for each reaction is given as below:

$$\begin{aligned} \frac{d[CH_4]}{dt} &= -AT^d[CH_4]^a[H_2O]^b \exp(-\frac{E}{RT}) \\ \frac{d[CH_4]}{dt} &= -AT^d[CH_4]^a[O_2]^b \exp(-\frac{E}{RT}) \\ \frac{d[H_2]}{dt} &= -AT^d[H_2]^a[O_2]^b \exp(-\frac{E}{RT}) \\ \frac{d[CO]}{dt} &= -AT^d[CO]^a[H_2O]^b \exp(-\frac{E}{RT}) \end{aligned} \tag{2}$$

The constant parameters of reaction rates are given in Table 1:

Table 1. CONSTANTS PARAMETERS OF REACTION RATES FOR JONES' MECHANISM

Reaction	A	d	a	b	E (cal/mol)
R1	0.30e09	0	1.0	1.0	30’000
R2	0.44e12	0	0.5	1.2	30’000
R3	0.68e16	-1	0.2	1.5	40’000
R4	0.275e1	0	1.0	1.0	20’000

Feed-forward Neural Network and Learning Algorithm

As shown in Fig. 1, a feed-forward multilayer neural network is a computing architecture that consists of massively distributed interconnected simple neurons [7]. For multilayer perceptron networks, the inputs propagate through the network, and the output of each neuron is evaluated according to:

$$y^n{}_i = f\left(\sum_{j=1}^{K_{n-1}} w_{ij}{}^n y_j{}^{n-1} + \theta_i^n\right)_{i=1,\ldots,K_n \,\&\, n=1,\ldots,N} \tag{3}$$

where y_i^n is the output of the i^{th} neuron of the n^{th} layer, $w_{ij}{}^n$ is the synaptic weight value of connection between j^{th} node of the $(n-1)^{th}$ layer and i^{th} neuron of n^{th} layer, and, finally, θ_i^n is the bias value of the i^{th} neuron of the n^{th} layer. The nonlinear activation function, $f()$, is differentiable and it must have a positive first derivative.

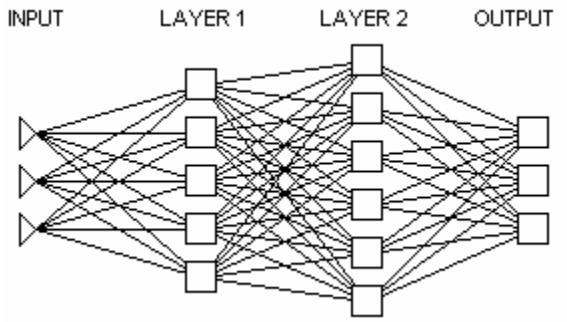

Figure 1. SCHEMATIC DIAGRAM OF A FEED-FORWARD MULTILAYER NEURAL NETWORK

In this work multilayer perceptron architecture, with two intermediate or hidden layers, has been employed. A hyperbolic-tangent function has been used as the transfer function. Figure 2 shows the neural network, used for modeling the mechanism. The inputs of the network are mole numbers of two main species CH_4 and O_2 and also two intermediate species CO and H_2 per unit mass at time t. The outputs are mole numbers of the same species per unit mass at the time $t+\delta t$. This neural network predicts the mass fractions of the this four reactive species and, therefore, the thermochemical state of the system at the end of the time step δt.

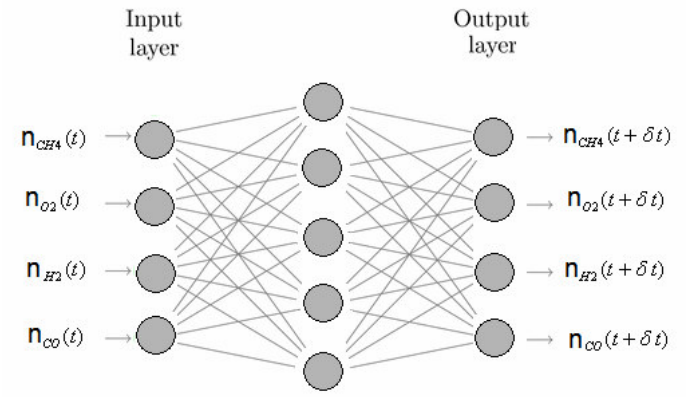

Figure 2. SCHEMATIC DIAGRAM OF USED NEURAL NETWORK

Figure 3 shows the synaptic weights between the neurons in different layers. For adjusting the weights, $w_{ij}^{\ n}$, in the network, we used the back-propagation method. This method is probably the most well-known and widely used learning algorithm that is based on gradient descent technique. This algorithm minimizes the value of the error function during the learning process [8]. The error function, E, is defined as:

$$E_t = \sum_{k=1}^{m}\sum_{i=1}^{n_o}(y_{i,k} - \hat{y}_{i,k})^2 \qquad (4)$$

where n_o is the number of output units, y_k and $\hat{y}_k$ are the expected and estimated outputs for the k^{th} learning pattern.

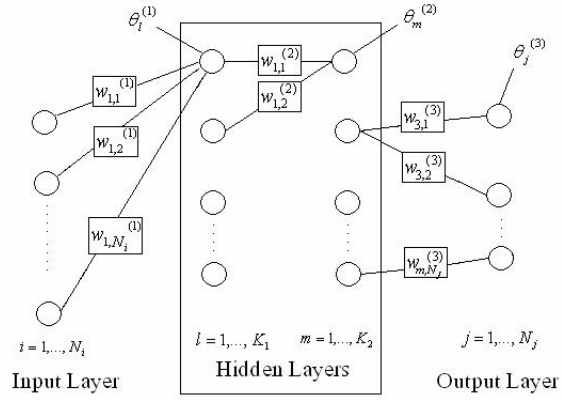

Figure 3. SYNAPTIC WEIGHTS FOR THE NEURAL NETWORK WITH TWO HIDDEN LAYERS

The back-propagation method adjusts the weights according to the following equation:

$$w_{ij}^{\ n}(t+1) = w_{ij}^{\ n}(t) + \eta\delta_j\hat{y}_i + \alpha[w_{ij}^{\ n}(t) - w_{ij}^{\ n}(t-1)] \qquad (5)$$

where t is the iteration number, and, $\eta > 0$ and $\alpha < 1$ are learning and momentum factors, respectively. These factors can be adjusted to speed up the convergence of the algorithm. Furthermore, the use of appropriate η and α reduces the possibility of being trapped in local minimums. The function δ_j represents the change in the error function of the j^{th} node with respect to the network inputs. It is:

$$\delta_j = -\frac{\partial E_t}{\partial\sum_i w_{ij}^{\ n}\hat{y}_i} \qquad (6)$$

The explicit form of δ_j depends on the activation function. It should be mentioned that j is an internal or an output node. The functional form of δ_j is given by:

$$\delta_j = \begin{cases} f'_j(y_j - \hat{y}_j) & j: output \quad node \\ f'_j\sum_i \delta_i w_{ij}^{\ n} & j: \text{int}\, ernal \quad node \end{cases} \qquad (7)$$

The function f'_j denotes the derivative of the activation function of the j^{th} node with respect to the total net inputs. It is:

$$f'_j = \frac{\partial \hat{y}_i}{\partial \sum_i w_{ij} \hat{y}_i} \tag{8}$$

Generating the training patterns

To define the thermochemical state of the system and generate the learning patterns for training of the neural network, one must first determine the scalars of the mechanism. Using the concept of mixture fraction, Chen et al. showed that a reduced mechanism, with seven reactive species, which has ten scalars (including seven reactive species, density, pressure and temperature), needs five known scalars to be defined [1]. They also showed that, these five scalars can not take independent values and they derived specific equations for the upper and lower bounds for each of these scalars.

For Jones' reduced chemical mechanism, there are six reactive species and, therefore, by adding temperature, pressure and density of the mixture, we have nine scalars. Using the concept of mixture fraction (defined as the normalized mass fraction of an atomic element originating from the fuel stream), the number of scalars can be reduced to three. Given a mixture fraction, three equations can be derived for atomic balance:

$$\begin{aligned} &H \quad atom: \quad 4n_{CH4} + 2n_{H2} + 2n_{H2O} = 4f / MW_{CH4} \\ &O \quad atom: \quad 2n_{O2} + n_{CO} + 2n_{CO2} + n_{H2O} = 2(1-f) / MW_{O2} \\ &C \quad atom: \quad n_{CH4} + n_{CO} + n_{CO2} = f / MW_{CH4} \end{aligned} \tag{9}$$

f is the mixture fraction, n_i indicates number of moles of species i per one kilogram of the mixture and MW_i indicates the molecular weight of species i.

Since this approach assumes that the enthalpy of the mixture is linear function of the mixture fraction, another equation is also added for the mixture enthalpy. Finally, considering ideal gas law for the mixture and constant pressure combustion, we will have six relations for thermochemical state of the system. Therefore, with the exception of mixture fraction, only three scalars are needed to determine the thermodynamic properties of this system. It is worthy to mention that n_{CH4}, n_{CO} and n_{O2} have been selected as these three scalars. Therefore, we are faced with four-dimensional domain $(f, n_{CH4}, n_{O2}, n_{CO})$ to define the system. For finding the look-up table computer storage requirement, this four-dimensional domain has been discretized into $(40,10,10,10)$ grid points. Mixture fraction is assumed to vary in the range of $0 < f < 0.6$. Around the stoichiometric point of methane and pure oxygen reaction ($f_{st.} = 0.2$) much closer and smaller grid points have been selected because the outputs of the network show much more non-linear behavior around this point.

Optimal Neural Network

After defining the learning patterns, the neural network must be trained by a number of input-output data pairs. The number of training sets must be large enough to be distributed in the whole range of the thermochemical state. 4000 sets have been selected for the training phase. The outputs of these sets have been found by a direct numerical integration process. The thermochemical database of CHEMKIN has been also used to compute the density and temperature of the system in each time step.

In the training phase of a neural network, it is important to check the behavior of the network for the unseen data within the thermochemical state domain. 3000 unseen pairs of input-output data have been selected for testing the network. These pairs are called test sets.

In order to find the optimal neural network architecture, number of hidden layers and hidden units, in each layer, has been increased and the total error of the network has been reported in Table 2. As can be seen, the neural network, with two hidden layers and 16 neurons in each layer, seems to be a good candidate. We also checked a neural network with two hidden layers and 22 units in each layer but, regarding the noticeable difference between the learning times and the small improvement in the error, the network with 2×16 hidden units was selected.

Table 2. ERRORS OF DIFFERENT NEURAL NETWORK ARCHITECTURES

	Hidden Neurons			
	8	12	16	16×2
Training Set Error	0.072	0.051	0.037	0.013
Test Set Error	0.108	0.077	0.059	0.032

RESULTS

Figure 4 shows the errors detected by the neural network for three species (methane, oxygen and carbon monoxide) as function of mixture fraction. As can be seen, the maximum errors for methane and oxygen take place around the

stoichiometric point but for the intermediate species, carbon monoxide, this maximum error takes place somewhere else.

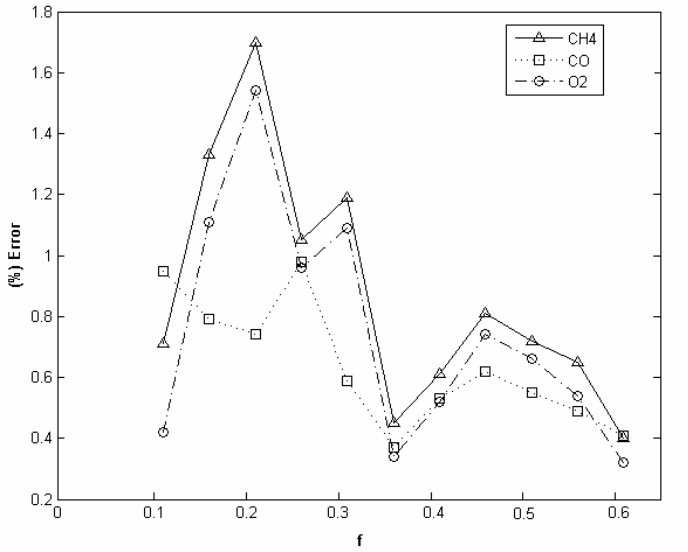

Figure 4. ERRORS FOR THREE SPECIES AS FUNCTION OF MIXTURE FRACTION

Figures (5) and (6) show the time evolution of methane and oxygen for two different values of mixture fractions: (A) near-stoichiometric point behavior (solid line for direct integration and dots for the neural network prediction), and, (B) a random selection of mixture fraction and other scalars of the system in the rich zone of the mixture, when $f > f_{st}$ (dotted line for direct integration and multiplication sign for neural network prediction).

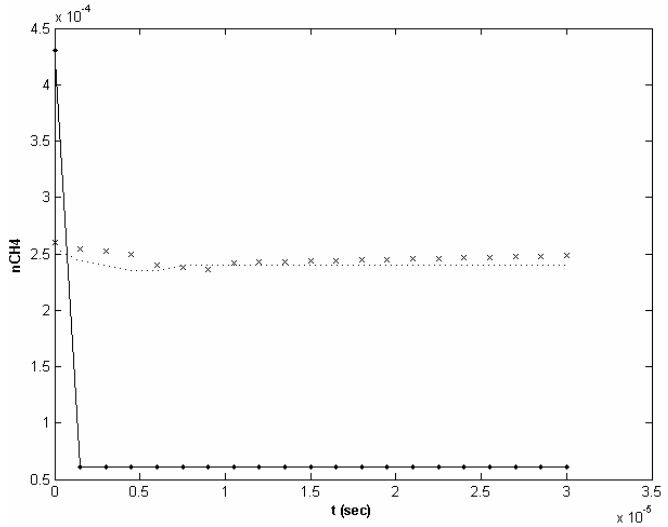

Figure 5. TEMPORAL CHANGES OF METHANE IN TWO DIFFERENT MIXTURE FRACTIONS: (A) NEAR STOICHIOMETRIC BEHAVIOR (SOLID LINE AND DOTS WHICH ARE CORRESPONDING TO THE DIRECT INTEGRATION METHOD AND NEURAL NETWORK MODELING, RESPECTIVELY) (B) A RANDOM SELECTION OF THE SCALARS IN THE RICH ZONE (DOTTED LINE FOR DIRECT INTEGRATION AND MULTIPLICATION SIGN FOR NEURAL NETWORK PREDICTION)

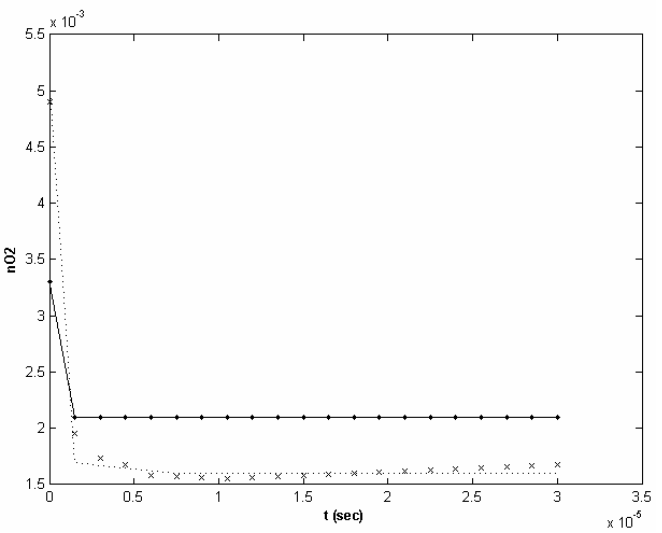

Figure 6. TEMPORAL CHANGES OF METHANE IN TWO DIFFERENT MIXTURE FRACTIONS: (A) NEAR STOICHIOMETRIC BEHAVIOR (SOLID LINE AND DOTS WHICH ARE CORRESPONDING TO THE DIRECT INTEGRATION METHOD AND NEURAL NETWORK MODELING, RESPECTIVELY) (B) A RANDOM SELECTION OF THE SCALARS IN THE RICH ZONE (DOTTED LINE FOR DIRECT INTEGRATION AND MULTIPLICATION SIGN FOR NEURAL NETWORK PREDICTION)

Once again, figure 7 shows the temporal evolution for the intermediate species, carbon monoxide, for two different regimes: (A) near stoichiometric point behavior (solid line for direct integration and dots for the neural network prediction), and, (B) a random selection of mixture fraction and other scalars of the system in the lean zone of the mixture, when $f < f_{st}$ (dotted line for direct integration and multiplication sign for neural network prediction). As can be seen, for the latter case, neural network prediction is a little higher than the actual direct integration values. In Fig. 7, solid line and dots are corresponding to the direct integration method and neural network modeling, respectively.

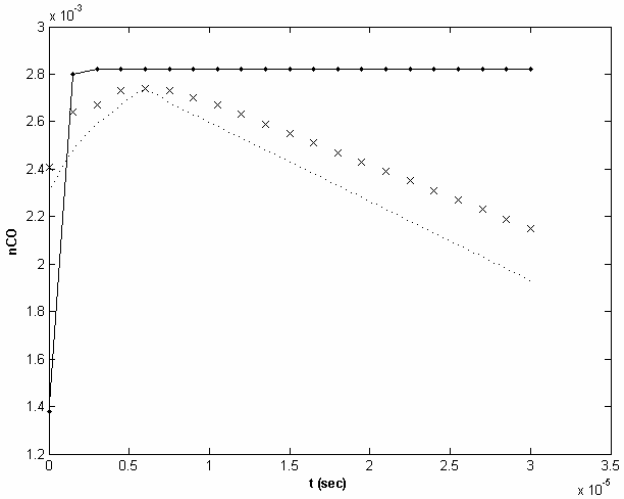

Figure 7. TEMPORAL CHANGES OF THE INTERMEDIATE SPECIES, CARBON MONOXIDE IN TWO DIFFERENT MIXTURE FRACTIONS: (A) NEAR STOICHIOMETRIC BEHAVIOR (SOLID LINE AND DOTS WHICH ARE CORRESPONDING TO THE DIRECT INTEGRATION METHOD AND NEURAL NETWORK MODELING, RESPECTIVELY) (B) A RANDOM SELECTION OF THE SCALARS IN THE LEAN ZONE (DOTTED LINE FOR DIRECT INTEGRATION AND MULTIPLICATION SIGN FOR NEURAL NETWORK PREDICTION)

COMPUTATIONAL COSTS

In order to make a comparison between neural network approach and one conventional method, like look-up tables, from the computational costs point of view, an important parameter must be calculated for each of these methods [5]. This parameters is the required memory storage factor, S. The required memory storage for look-up table method in bytes is given by:

$$S_{LUT} = 4N_J N_\varepsilon N_t (N_1 N_2 ... N_J) \qquad (10)$$

where N_J is the number of reactive scalars, N_ε is the number of discretized nodes of mixture fraction, and N_i for i=1,2,..,N is the number of tabulated scalars. N_t is the number of reaction time intervals.

The required computer storage for neural network model, in bytes, is given by:

$$S_{NN} = 4N_t[(N_l + 1)K_l + \sum_{l=1}^{L-1} K_{l+1}(K_l + 1) + (N_J + 1)K_L] \qquad (11)$$

where K_l is the number of neurons in the l^{th} hidden layer, N_I and N_J are the number of input and outputs of the network.

For Jones' reduced chemical mechanism, with five scalars, $(f, n_{CH4}, n_{O2}, n_{CO})$ which has been discretized into $(40,10,10,10)$ grid points and a neural network with four inputs, four outputs and two hidden layers with sixteen neurons, in each layer, the required computer storage has been calculated for both methods, look-up table and neural network method. The relevant results are shown in Table 3.

Table 3. COMPARISON BETWEEN COMPUTER STORAGE REQUIREMENT OF LOOK-UP TABLE METHOD AND NEURAL NETWORK METHOD FOR FIVE TEMPORAL INTERVALS (BYTES) AND THEIR RATIO

	Required computer storage (bytes)	Ratio
Look-up table	64e+05	740
Neural network	8640	1

CONCLUSION

In this research, an important reduced chemical mechanism of methane/oxygen reaction has been successfully modeled by an artificial neural network with considerably decreasing of the computational costs. Also the approach for building the neural network architecture and choosing the optimal network has been stressed. There is good agreement between neural modeling and direct integration for the main species, like methane and oxygen, but for an intermediate species, like carbon monoxide, there are some inconsistencies between the prediction of the network and direct integration method.

The work is now under development to model other chemical mechanisms of methane/air and methane/oxygen combustion, like Srivatsa mechanism [9], skeletal mechanism [10], and full mechanism (if possible), Afterwards, we will have the possibility to provide the detailed comparison between the neural modeling and computational costs of the mentioned mechanisms.

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