

Machine learning tabulation of thermochemistry for turbulent dimethyl ether (DME) flames

Anxiong Liu^{a,b}, Tianjie Ding^c, Runzhi Liu^b, Stelios Rigopoulos^{c,*}, Kun Luo^{a,b,*}

^aShanghai Institute for Advanced Study, Zhejiang University, Shanghai 201203, China

^bCollege for Energy Engineering, Zhejiang University, Hangzhou, 310013, China

^cDepartment of Mechanical Engineering, Imperial College London, London SW7 2AZ, United Kingdom

Abstract

The present work applies a machine learning tabulation methodology for the thermochemistry of dimethyl ether (DME), a typical biodiesel fuel, to accelerate the real-time computation of a large-size DME mechanism in turbulent non-premixed combustion. This approach is known as the hybrid flamelet/random data and multiple multilayer perceptrons (HFRD-MMLP) method (Ding et al., 2021). The essence of the HFRD-MMLP method lies in the generation of training data using the HFRD approach, which enhances the capacity of generalisation for the reactive composition space encountered in practical turbulent combustion by using the random process to expand the training dataset from laminar flamelets. The MMLP artificial neural networks (ANNs) are then trained to predict different composition states, aiming to improve the accuracy of ANN predictions. To validate the effectiveness of the ANNs, they are initially tested on 1-D laminar flame simulations with varying strain rates. Subsequently, a test is conducted on Sandia non-premixed turbulent DME series flames D and F, with an increasing jet Reynolds Number. The results regarding species mole fractions and temperature show overall excellent agreement with the direct integration method. The HFRD-MMLP method achieves speed-up factors of over 16 and 10 for the reaction step, and total computational costs, respectively, in the LES-PDF simulation with the DME mechanism in this work, surpassing the speed-up factors of approximately 12-14, and below 5 for the reaction step, and total time costs, respectively, in previous works on GRI-1.2 mechanism for CH₄ (Ding et al., 2021) and CH₄/H₂ (Ding et al., 2022) combustion. This indicates that the HFRD-MMLP method is highly effective for mechanisms of large size in reducing the computational cost by avoiding a significant number of highly stiff ordinary differential equations (ODEs) integration. This approach can be applied in real-time calculations of reaction source terms in methods including Direct Numerical Simulation (DNS), Probability Density Function (PDF) methods, unsteady flamelet, Conditional Moment Closure (CMC), Multiple Mapping Closure (MMC), Linear Eddy Model (LEM), Thickened Flame Model, the Partially Stirred Reactor (PaSR) method (as in OpenFOAM) and laminar flame computation.

Keywords: Turbulent non-premixed flames, Machine learning, Artificial neural networks (ANNs), chemistry tabulation, biodiesel fuels

1. Introduction

The escalating consumption of fossil fuels is imposing substantial pressure on air quality, necessitating the exploration of alternative fuels that are both sustainable and environmentally friendly. Dimethyl ether (DME, C_2H_6O) has emerged as a promising renewable fuel source, characterised by a high cetane number and excellent evaporation properties, presents a viable option to replace diesel in compression-ignition engines [1]. Furthermore, owing to its unique oxygenated molecular structure, which lacks direct carbon-carbon bonds, DME has the potential to significantly mitigate emissions of carbon monoxide (CO), nitrogen oxides (NO_x), and soot [2]. DME can be sourced from an array of feedstocks, such as natural gas, crude oil, residual oil, coal, waste products, and biomass, and thus replacing diesel with DME can considerably reduce the demand for oil. The efficacy of DME as an alternative fuel has been investigated in a number of research studies, and comprehensive reviews of these works are available in references [1–3].

CFD simulations for turbulent reactive flow, particularly those involving DME fuel, usually require detailed chemical mechanisms and real-time computation of the chemical source term. Various simulation methods, such as Direct Numerical Simulation (DNS), Probability Density Function (PDF) methods, unsteady flamelet, Conditional Moment Closure (CMC), Multiple Mapping Closure (MMC), Linear Eddy Model (LEM), Thickened Flame Model, the Partially Stirred Reactor (PaSR) method (as in OpenFOAM) and laminar flame computation (see e.g. Refs. [4–6]), require direct coupling of chemistry and flow. However, these methods face challenges due to the significant CPU time required for computing the chemical kinetics through real-time direct integration of a large number of stiff ordinary differential equations (ODEs). This computation is necessary at every grid node and for each time step of the simulation. The high computational cost of the chemical kinetics computation poses a bottleneck for these simulations. For example, in LES-PDF simulations using Eulerian stochastic fields for turbulent diffusion flames, the direct integration of the reaction step alone can account for 70% to 95% of the total CPU time cost, depending on the size and stiffness of the mechanism in typical cases. It limits the feasibility of applying these methods to real-world engineering problems. Therefore, researchers are constantly exploring strategies to mitigate this issue and make these simulation methods more efficient and practical.

There are two main approaches to accelerate the real-time calculation of the chemical kinetics. The first involves utilising mathematical techniques to reduce the original kinetics using a simplified skeletal mechanism. These techniques typically include sensitivity analysis [7], direct relation graph (DRG)[8, 9], pathway flux analysis (PFA) [10, 11], Computational Singular Perturbation (CSP) [12], Rate-Controlled Constrained Equilibrium (RCCE) [13, 14], global path selection (GPS) [15], differential evolution algorithms with strategy adaption (SaDE) [16], genetic algorithms (GA) [17, 18] and particle swarm optimisation algorithm (PSO) [19]. The reduced chemical kinetics obtained through the above mathematical methods are usually validated in the 0D perfectly stirred reactor, 1D premixed and

*Corresponding author

Email address: zjulk@zju.edu.cn (Kun Luo)

counterflow diffusion flames. While key flame parameters of the reduced mechanisms, such as the chemical reaction time scale, flame propagation speed, and concentrations of major species in the mixture fraction space or the progress variable space, agree well with the detailed mechanisms, information about minor species is often disregarded, despite its importance sometimes. For example, emissions such as NO_x and soot are crucial aspects of combustion. Detailed mechanisms that include NO_x (GRI 3.0) [20] or PAHs [21–23] are commonly employed to predict NO_x [24] and soot emissions [25, 26]. However, if the chemical pathways of NO_x or PAHs are retained, it becomes challenging to reduce the detailed mechanisms into smaller ones. This is because the NO_x and PAHs are essentially minor species but their pathways are interconnected with almost all other species in detailed mechanisms. Consequently, in certain scenarios, extracting a skeletal mechanism using the aforementioned mathematical methods may not be an effective choice. Moreover, even the number of species is reduced by half or further, the real-time direct integration of chemical kinetics remains computationally expensive.

The second option is to accelerate the real-time direct integration of the detailed chemical kinetics by utilising various forms of tabulations, which is a pre-computed database of reaction source terms. The Look-up Table (LUT) [27] is the earliest and most straightforward method where interpolation is directly performed within the pre-computed database. In order to reduce the memory requirements and facilitate the application to large-size mechanisms, two alternative techniques have been developed: Intrinsic Low-Dimensional Manifolds (ILDM) [28] and the In-Situ Adaptive Tabulation (ISAT, where the chemistry tabulation is updated during the simulation) [29]. Another approach is to employ artificial neural networks (ANNs) for chemistry tabulation, which are trained machine learning models capable of approximating highly non-linear to the real-time integration of chemical kinetics. Compared to traditional tabulation approaches, ANNs require lower memory storage and offer faster computational speed.

Early research by Christo et al. [30, 31] and Blasco et al. [32] utilised a single ANN to tabulate simple mechanisms, specifically, 3-step and 4-step combustion chemical systems. However, as the number of species and reactions increases, tabulating the exponentially expanding composition space with a single ANN became challenging. Consequently, multiple ANNs were introduced to cover the entire composition space of complex mechanisms. Blasco et al. [33] and Chen et al. [34] divided the composition space into several subdomains based on mixture fraction or temperature, and each subdomain was fitted with an individual ANN. Subdomain clustering was facilitated using the Self Organising-Map (SOM) [35–37]. Another approach involving multiple ANNs was to predict each reactive scalar with an individual ANN [38, 39], significantly improving the accuracy of ANN predictions. Deep learning approaches (employing ANNs with more than three layers), were also trained with reactive scalars to predict chemical sources [40, 41]. Furthermore, ANNs have also been widely applied in the non-real-time integration of chemical kinetics approaches, *e.g.* the steady flamelet model, which employs mixture fraction or progress variables to tabulate all reaction variables. For instance, Kempf et al. [42] trained individual multi-layer perceptrons (MLPs) for each predicted flamelet variables to conduct LES simulations of a non-predicted piloted methane-air jet flame. Similarly, Hansinger et al. [41] utilised a deep ANN to tabulate flamelet/progress variables and applied in LES simulations of the Sandia/Sydney flame. However, flamelet/progress variable models do not impose the same high demands on

CPU times as the chemistry real-time integration, making ANN tabulation less essential for non-real-time chemistry computations.

In practice, the use of ANNs chemistry tabulation for accelerating the computation of chemical source terms encounters a major challenge: ANNs should ideally possess strong generalisation capabilities, enabling their application to a wide range of realistic combustion problems. The generation of appropriate training data is a crucial step in most machine learning scenarios, including ANN chemistry tabulation, because it directly influences the effectiveness and generalisation performance of ANNs. The training data must encompass the entire composition space of reactive scalars encountered in realistic combustion problems. Moreover, it is preferable to generate a large quantity of training data using simple and time-saving approaches, rather than obtaining it from complex turbulent combustion scenarios where the direct integration of chemical source terms is computationally expensive. Sen and Menon [43, 44] generated datasets through laminar flame-vortex interaction simulations and stand-alone linear edge mixing (LEM) simulations [45]. Chatzopoulos and Rigopoulos [36] and Franke et al. [37] employed unsteady laminar flamelet simulations for data generation, while Wan et al. [40] used a canonical stochastic micromixing problem. An et al. [46] collected training data from a RANS simulation of the hydrogen/carbon monoxide/kerosene mixture in a rocket-based combustor and subsequently applied ANNs to LES simulation.

Readshaw et al. [39], Ding et al. [47, 48] have recently proposed a hybrid flamelet/random data (HFRD) method based on unsteady laminar flamelet sampling method developed by Chatzopoulos and Rigopoulos [36], Franke et al. [37]. In this approach, they initially utilise the laminar flamelet dataset to ensure the validity and meaningfulness of the physically thermochemical states. Subsequently, the random process is employed to expand the training dataset beyond the limits of flamelet solutions, thus encompassing the composition regimes that are relevant to complex turbulent combustion problems. The results demonstrated that the HFRD method generates datasets that allow for considerable capacity for generalisation. These applications encompass turbulent non-premixed flames using methane fuel (Sandia D,E and F flames [47], Sydney flame L [39]), as well as methane/hydrogen blends (Sydney flame HM1), and turbulent premixed flame. Another technique in these studies is the utilisation of the multiple multilayer perceptron method (MMLP) to enhance the accuracy of ANN predictions. Herein, multiple MLPs are trained for individual reactive scalars across different concentration regions.

To the author's knowledge, great progress of ANN chemistry tabulation have primarily been limited to small-scale chemical mechanisms, such as the GRI-1.2 mechanism [49]. This mechanism involves 31 species and 171 chemical reactions, making it suitable only for modelling the combustion of small-molecule hydrocarbons. However, the complexity of DME (dimethyl ether) detailed mechanisms surpasses that of GRI-1.2 due to the inclusion of chemical pathways for oxygenated molecules in addition to hydrocarbons. For instance, Fischer et al. [50] developed a comprehensive mechanism consisting of 82 species and 351 reactions, while Zhao et al. [51] proposed a mechanism involving 55 species and 290 reactions. Furthermore, Burke et al. [52] developed an extensive mechanism comprising 113 species and 710 reactions. The real-time calculation of DME mechanisms in turbulent combustion simulations will result in a quadratic growth of time and stiffness. Therefore, it becomes imperative to apply the ANN methodol-

ogy to DME fuel, enabling accelerated combustion simulations that are more economically feasible.

In this paper, we apply the HFRD-MMLP methodology to tabulate the DME mechanism developed by Zhao et al. [51]. The use of DME fuel serves as a robust validation to reaffirm and extend the HFRD-MMLP method to more intricate mechanisms and fuels. This work is structured as follows: Section 2 outlines the methodology, focusing on acquisition of training data from laminar diffusion flamelets and then augmented by the random process for better generalisation. Section 3 shows *a priori* test of the ANNs into laminar flamelet simulations. We show the performance comparisons between the selected ANNs trained with the HFRD-MMLP method and the ANNs based on laminar flamelet dataset but without the random augmentation. Additionally, we demonstrate the accuracy enhancement achieved through using the multiple MLPs for each reactive scalar. Moving forward, in Section 4, the best performing ANNs is applied to the LES-PDF stochastic simulations of the Sandia DME flame series, and the CPU time breakup are also discussed for the ANNs simulation. Finally, we conclude the paper by summarising the key findings.

2. Tabulation of chemistry using the methodology of hybrid flamelet/random data and multiple multilayer ANNs

2.1. Overview of the ANN tabulation process

The reaction source term of a chemical species is expressed by the following ordinary differential equations (ODEs),

$$\dot{\Omega}_i = \frac{dy_i}{dt} = f_i(\mathbf{Y}, h, p), \quad i = 1, 2, \dots, n \quad (1)$$

where y_i is the concentration of species i , and $\mathbf{Y} = (y_1, y_2, \dots, y_n)$ is the species composition space. h is specific enthalpy (including the sensible enthalpy and the enthalpy of formation) of mixture which is a constant value during the reaction time step t . For reactions under a fixed pressure p , the species concentration change Δy_i can be expressed as a function of the initial states $\mathbf{Y}$ and h in a constant time step Δt :

$$\Delta y_i = F_i(\mathbf{Y}, h), \quad i = 1, 2, \dots, n \quad (2)$$

where F_i is a function that is implicitly determined by the solution of the chemical kinetics. ANNs are a class of machine learning models to fit the nonlinear functions $F_i(\mathbf{Y}, h)$, replacing the need of the direct integration of chemical kinetics. In this study, the multilayer perceptron (MLP), a widely used ANN model for nonlinear function fitting, is utilised. An MLP consists of two hidden layers comprising 50 neurons, facilitating accurate predictions of the species concentration change Δy_i . The DME mechanism by Zhao et al. [51] contains 55 species, with Ar and He excluded from participating in any chemical reactions, resulting in a reduced input dimensionality of 53 species and the specific enthalpy h . Therefore, each MLP takes 54 inputs and generates a single output denoting the species concentration change Δy_i . Figure 1 illustrates the architecture of an MLP, featuring an input layer, two hidden layers and an output layer.

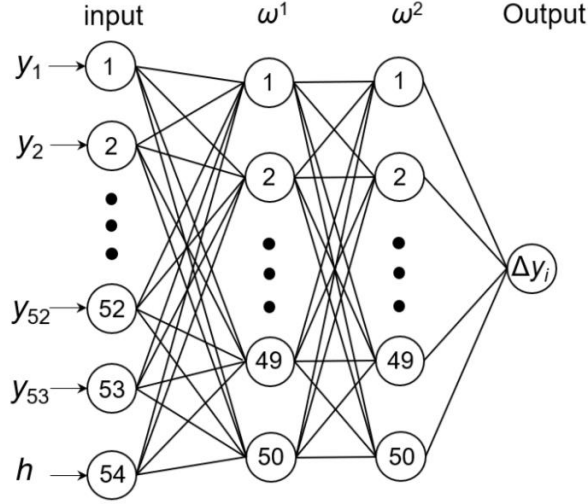


Figure 1: Architecture of the multilayer perceptron (MLP)

The output of each layer for the MLP can be calculated using the following equations:

$$a_j^1 = f^1 \left(b_j^1 + \sum_{i=1}^N \phi_i w_{i,j}^1 \right) \quad N = 54, \quad j = 1, 2, \dots, M_1 (M_1 = 50) \quad (3a)$$

$$a_j^2 = f^2 \left(b_j^2 + \sum_{i=1}^{M_1} a_i^1 w_{i,j}^2 \right) \quad M_1 = 50, \quad j = 1, 2, \dots, M_2 (M_2 = 50) \quad (3b)$$

$$o = f^3 \left(b^3 + \sum_{i=1}^{M_2} a_i^2 w_i^3 \right) \quad M_2 = 50 \quad (3c)$$

The superscript indicates the layer and the subscript j indicates the index of a neuron within the layer. The inputs are $\phi_i = [y_1, y_2, \dots, y_{52}, y_{53}, h]_{\text{T}}$. The weights $w_{i,j}$ of the j^{th} neuron in the present layer is connected with the i^{th} neuron of the previous layer. The inputs go forward through each layer using the above equations until the final output $o(\Delta y_i)$ is calculated. In this work, the hyperbolic tangent function and the linear function are employed as the activation functions of the hidden layers $f^1(x)$, $f^2(x)$ and the output layer $f^3(x)$, respectively.

The ANNs initially need to undergo training with a designated dataset. Throughout this training phase, the ANN's parameters are fine-tuned to align with a specific input and output data set. In this study, all ANNs are trained using the MATLAB neural network toolbox, and the chosen training algorithm is Bayesian Regularisation [53, 53, 54]. The Multi-Layer Perceptron (MLP) structure and training algorithm settings remain consistent with those utilised in the earlier research [39, 47].

2.2. Hybrid flamelet/random data (HFRD) generation approach

The dataset obtained through the flamelet sampling method can broadly cover the composition space expected in turbulent combustion problems. The previous works [36, 37] proposed a canonical method to generate data from

an ensemble of flamelets to represent various combustion scenarios, including non-premixed laminar and turbulent flames. However, it is still limited to composition states that are only generated during flamelet simulations and thus biased towards realistic combustion states. Figure 2 illustrates that flamelet sampling is unable to fully cover the range of combustion states in Sandia DME flames (the sampling procedures of the flamelet datasets and the numerical setup for the Sandia DME flames will be described in detail in the following sections). For instance, in the composition space of OH and CO₂ (Fig. 2a), the flamelet dataset has significantly fewer samples in the region of [0.001, 0.003] for CO₂ and [1e-4, 3e-4] for OH compared to the Sandia DME turbulent flames. Moreover, the plot of mixture fraction z and the specific enthalpy h (Fig. 2b) reveals evident streak patterns that in the flamelet dataset, while the Sandia flame data lies outside the scope of flamelet representation. Herein, the hybrid flamelet/random data (HFRD) generation approach to achieve greater generality was proposed in the previous works [39, 47] and applied in the data generation of the present work for the DME mechanism.

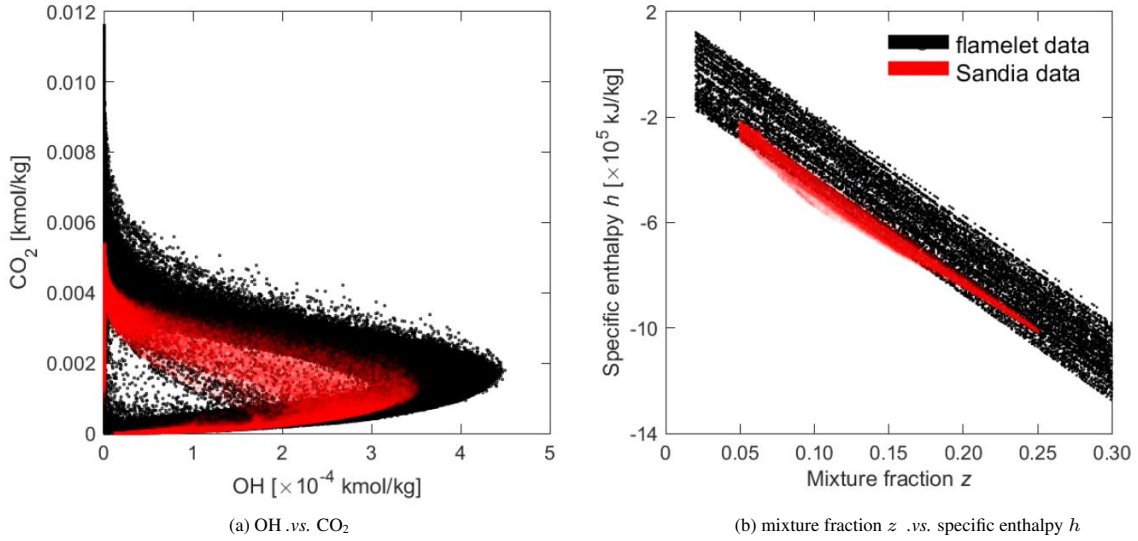


Figure 2: Scatter plots comparing the composition space of the flamelet data and the data collected from the LES-PDF simulations of the DME Sandia flames

For the first step, a series of 1-D laminar counterflow diffusion flamelet simulations are performed to collect data

$$\rho \frac{\partial y_i}{\partial t} = \rho \frac{\chi(z)}{2} \frac{\partial^2 y_i}{\partial z^2} + \dot{\omega}_i \quad (4)$$

where z is mixture fraction, y_i is the species concentration (kmol/kg) and $\dot{\omega}_i$ is the reaction source term of the i th species, and χ is the scalar dissipation rate given by the following equation

$$\chi(z) = \frac{S}{\pi} \exp \left[-2 \left(\text{erfc}^{-1}(2z) \right)^2 \right] \quad (5)$$

where S is the strain rate. By varying the strain rate and the mixture, a wide range of composition states from equilibrium to extinction can be collected. In this work, a total of 200 flamelet simulations were conducted, with the

minimum and maximum strain rates set to 1 s^{-1} and 2200 s^{-1} , respectively. To expand the range of specific enthalpy covered by the training data, the initial temperatures of the fuel and air streams were randomly selected between 150 K and 350 K. The composition states were collected within the mixture fraction flammability range from 0.03 to 0.3 and a minimum combustion temperature of 500 K to ensure effective reactivity. Overall, approximately 840,000 data samples were collected to yield the above physical reasonable conditions.

The next crucial step is to expand the generality of the flamelet dataset using the random data approach. For each state (h and y_i) sampled from the flamelet dataset, a new random state can be generated using the following equations:

$$h_{\text{rnd}} = h + \frac{c}{a} (h_{\text{max}} - h_{\text{min}}) \quad (6a)$$

$$y'_i = y_i^{(1+\frac{c}{b})} \quad (i \neq i_{\text{N}_2}) \quad (6b)$$

$$y'_{\text{N}_2} = y_{\text{N}_2} + \frac{c}{d} (y_{\text{N}_2, \text{max}} - y_{\text{N}_2, \text{min}}) \quad (6c)$$

$$y_i^{\text{rnd}} = \frac{y'_i}{\sum y'_i} \quad (6d)$$

Compared with previous works of the HFRD approach for GRI 1.2 mechanism [47, 48], the coefficients a , b , and d in this study are set to a larger value of 20 to make a smaller random range. This is due to the fact that the DME mechanism has more species than GRI-1.2, making it difficult to satisfy all constraints in Table 1 required if using a large random range. Apart from the basic constraints of the mixture fraction and minimum reacting temperature, the random data must have appropriate molar element ratios. In the combustion of DME/air mixture, the molar element ratios of H/C is fixed and is exactly 3.0 based on equal diffusivity assumption. But the element ration of O/N cannot be directly used to determine whether the random state is physically reasonable, because the element O originates from both the oxygenated fuel DME ($\text{C}_2\text{H}_6\text{O}$) and the air. To calculate the O/N ratio of air, the contribution of element O from DME should be deducted from the total concentration and the following equations are employed for element constraints,

$$C_{\text{ON}} = \frac{n_{\text{O}} - n_{\text{C}}/2}{n_{\text{N}}} \quad (7)$$

where n_{O} , n_{C} and n_{N} refer to the total concentration of the corresponding element, and this constraint of the air O/N ratio C_{ON} is equal to 0.2658. When applying ANNs, the conservation of elements cannot be fully guaranteed due to ANN prediction errors. Therefore, the element ratios in the training data should be set a wider range instead of fixed values to cover the slightly different element ratios in the ANN predictions. Table 1 shows the maximum and minimum values of the constraints for the random data. In this present work, a total of about 830,000 random data were generated.

Once the random dataset has been obtained, all data goes through a single reaction step using the direct integration with the VODE solver [55] (DVODE) to calculate the target outputs (the change of species concentration) with the time step $\Delta t = 10^{-6} \text{ s}$ for ANN training. The DVODE solver is also used for the flamelet and LES-PDF combustion

Variables	Minimum	Maximum
H/C ratio	2.9	3.1
C_{ON}	0.255	0.275
Temperature	500 K	–
Mixture fraction	0.03	0.3

Table 1: Constraints for DME random data generation

simulations.

Figure 3 shows that now the random data can completely cover the combustion states in Sandia DME flames. Compared with the flamelet data in Fig. 2, the random data is more uniformly distributed and covers a larger space. In addition, the streak patterns no longer appear in the scatter plot of mixture fraction .vs. the specific enthalpy (Fig. 3b), which means that the random data generation data method will endow the ANNs with greater generalisation and suitable for the realistic combustion problems.

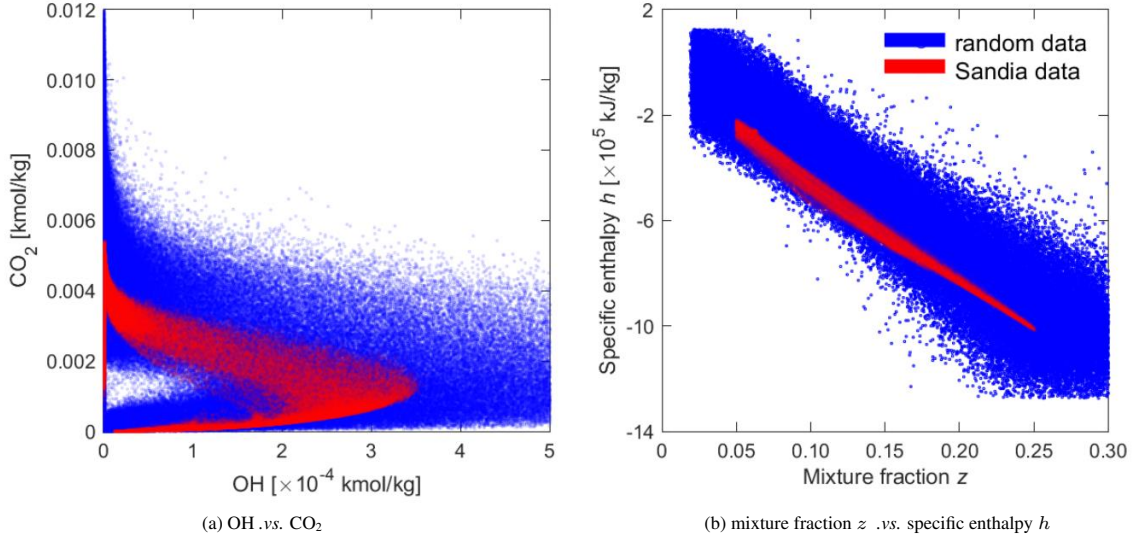


Figure 3: Scatter plots comparing the composition space of the random data and the data collected from the LES-PDF simulations of the DME Sandia flames

Once the training dataset has been generated, we employ the Bayesian regularisation method (see Sec. 2.2) to train the whole dataset. All the ANN training was conducted with the MATLAB neural network toolbox. Each ANN is trained for three times of 500 epochs, and each time of 500 epoch training uses 100,000 data samples, which are randomly chosen from the entire random dataset. Therefore, totally 300,000 data states from 830,000 random data take part in the training of each ANN. Now we compare the prediction results of the ANNs trained with flamelet data and random data, respectively, for the major species H, H₂O and CO in Fig. 2.2 and Fig. 5. The ANNs trained with flamelet data predict the flamelet output in good agreement with high correlation coefficients R_{flamelet} (the black scatters lies on the proportional function line $y = x$) because the training set is the same as the test set. However, when the test set is switched to the DME Sandia flame, the ANNs trained on flamelet data fail to provide correct target

outputs and the correlation coefficients R_{flamelet} are all below or around 80%, indicating a weakness in generalisation ability. In contrast, the trained ANNs using random data can predict accurately in both tests and correlation coefficients R_{random} are significantly improved, showing their improved generalisation capacity for the data samples from both flamelets and the LES-PDF simulation DME turbulent flames.

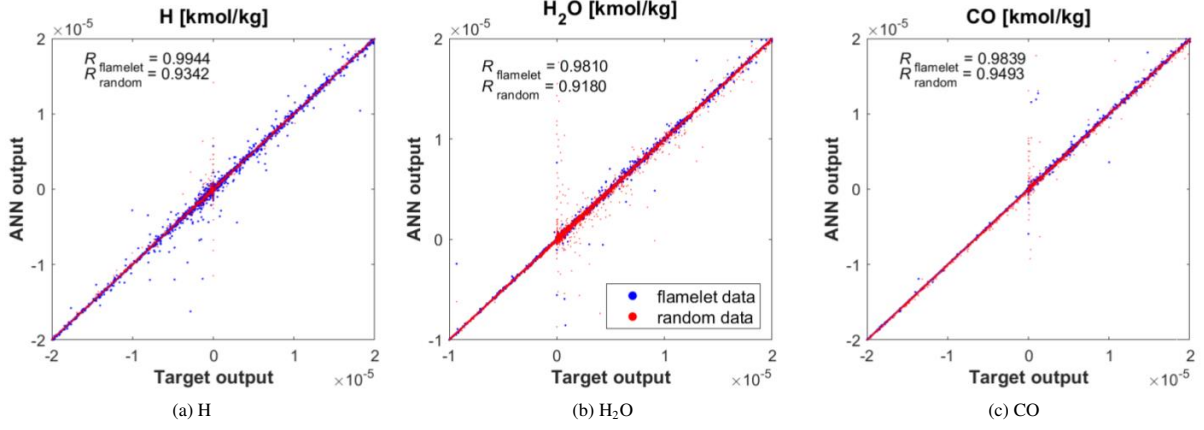


Figure 4: Comparison of predictions for the flamelet composition space using ANNs trained with the flamelet data and random data, respectively

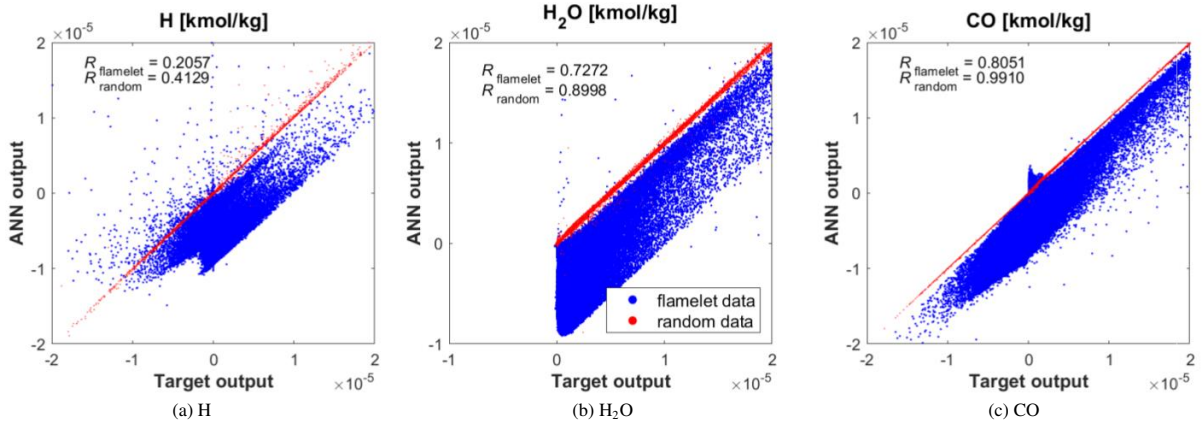


Figure 5: Comparison of predictions for the composition space of the LES-PDF simulated DME Sandia flame D using ANNs trained with the flamelet data and random data, respectively

2.3. Multiple MLPs (MMLPs) method

The multiple mutipayer perceptrons (MMLPs) approach proposed in the previous work [47] is also applied in this study. The motivation of introducing MMLPs is that using only a single MLP for the whole dataset results in absolute errors of a similar magnitude for all data, causing larger relative errors for data with small output magnitude. Therefore, a separate MLP is trained for data to adjust errors with the output of small concentration changes during reaction and thus improve the prediction accuracy.

For example, for the output range of a target species $[-k, k]$, the MLP absolute error may be of the order of 1% of k . For the output value is within the range $[-k/10, k/10]$, the relative error becomes 10%. At the moment, a separate MLP is trained using the random data within the small range and its performance is expected to be greatly improved compared with the first MLP trained with the entire random dataset. If the relative errors predicted by the second MLP are still large for data within further smaller output values (*e.g.* $[-k/100, k/100]$), a third MLP will be trained in such data range to improve the prediction accuracy. Finally, for each species, multiple MLPs are trained, with different magnitudes of species concentration changes. Based on the MLP performance of outputs, the output range for each new MLP is selected as 1/10, 1/5 or 1/4 of the previous range.

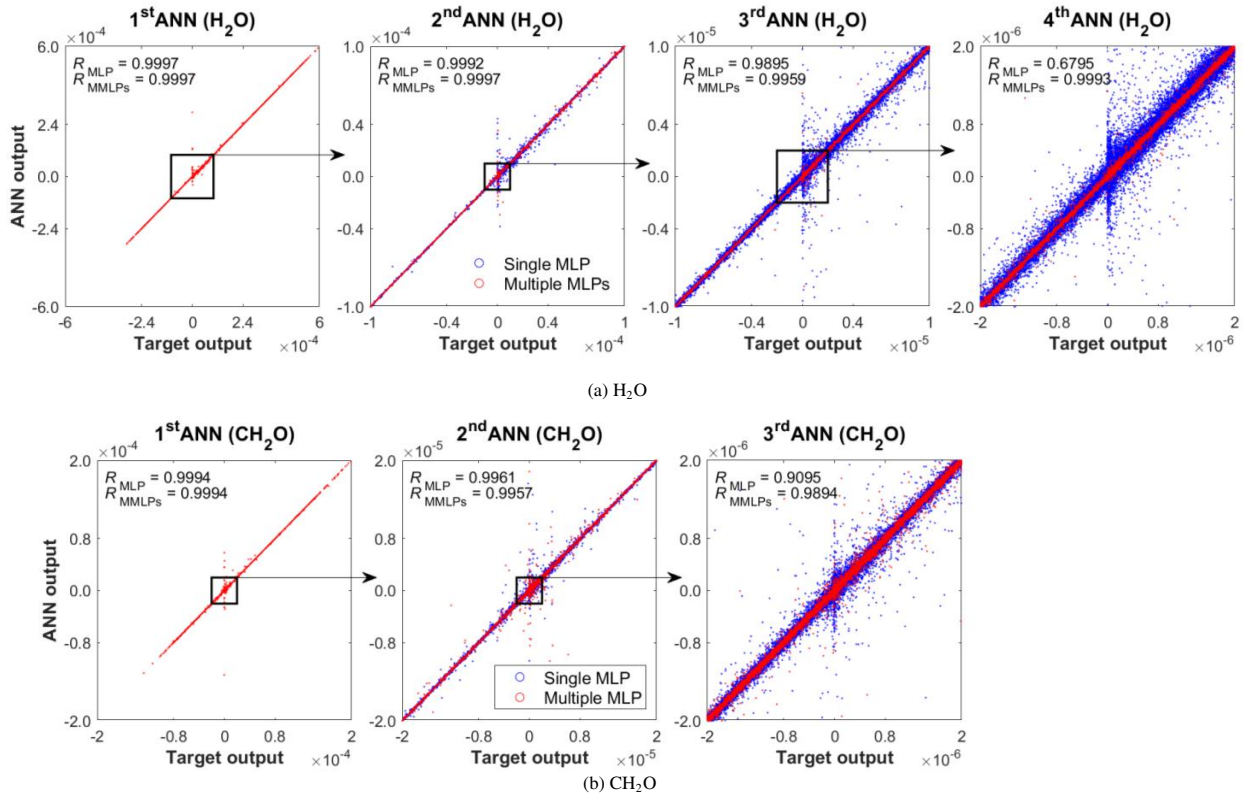


Figure 6: Comparison of predicted outputs for the training set of the random data using a single MLP and multiple MLPs (MMLPs)

Figure 6 displays the predicted outputs for the training set of the random data using a single MLP and multiple MLPs (MMLPs) networks. As the first MLP of the trained MMLPs is exactly the single MLP, so that the predicted outputs by the single MLP is thoroughly overlapped by the scattered predicted outputs by the first MLP of the MMLPs covering the broadest range. As the data range is narrowed, the results by the multiple MLPs are more concentrated on the diagonal line $y = x$ while the single MLP method results in more dispersed points, which means the multiple MLPs approach has substantially improved the prediction accuracy. The correlation coefficients for MMLPs are significantly improved.

Figure 7 quantitatively shows the multiple MLPs performance denoted by the relative RMSE(Root Mean Square

of Errors) of species CH_3 , O_2 , H_2O_2 and DME over the test data of the flamelet data, compared to the relative RMSE using only one single MLP. The relative RMSE is defined as

$$\text{relative RMSE} = \frac{1}{N_d} \sqrt{\sum_{q=1}^{N_d} \frac{(s_q - o_q)^2}{|s_q + \epsilon|}} \quad (8)$$

As the data range is narrowed, the relative RMSE with one single MLP increases, while the relative RMSEs with multiple MLPs for corresponding data ranges have dropped by 0.5 - 2 orders of magnitude. This means that the multiple MLPs method has significantly improve the accuracy.

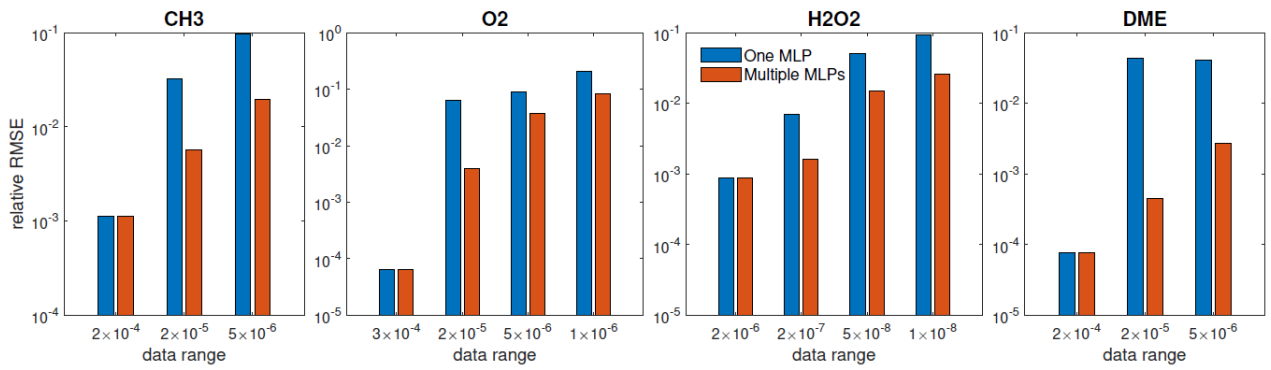


Figure 7: Comparison of the relative RMSEs for the test set of the flamelet data using a single MLP or multiple MLPs (MMLPs)

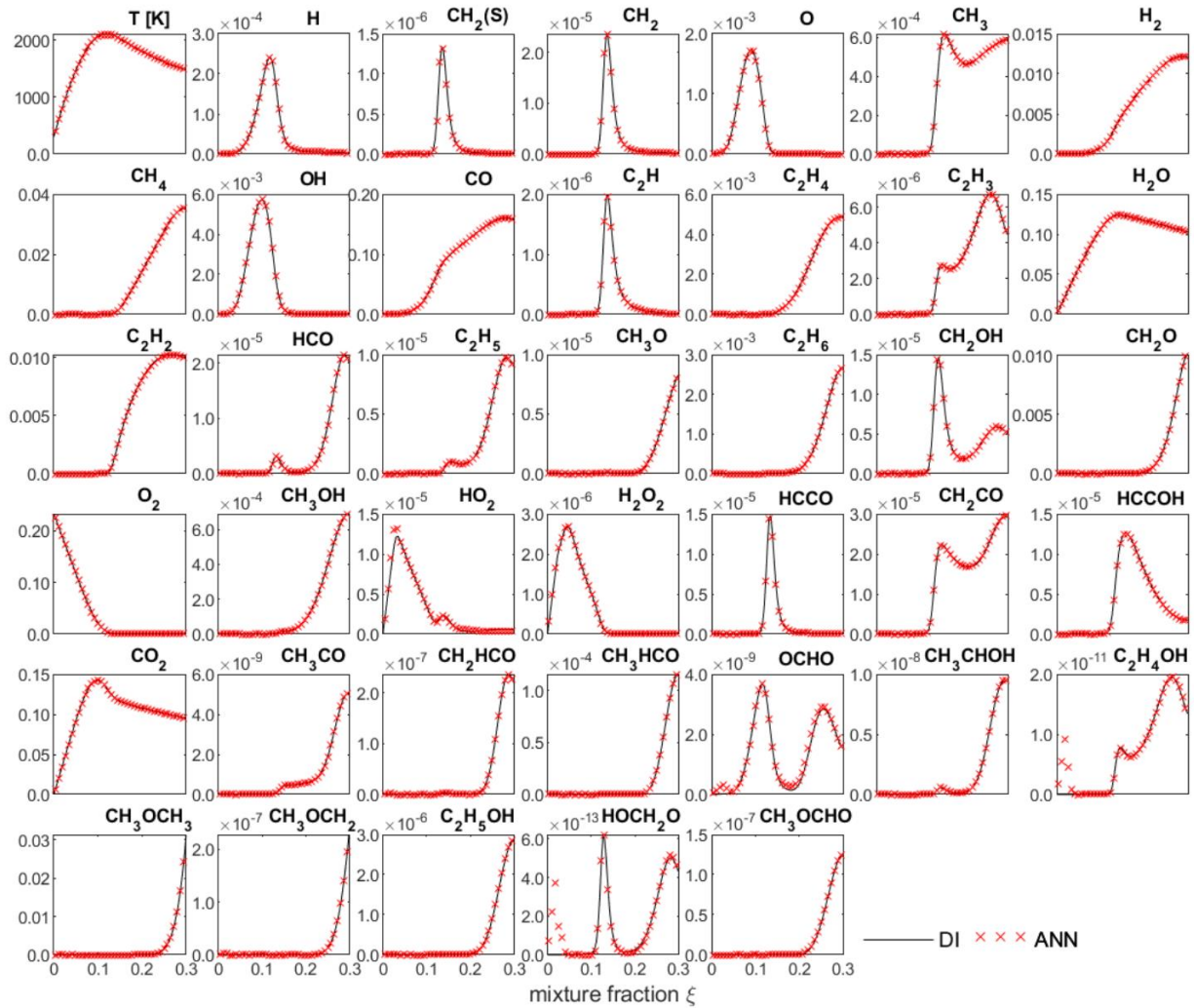
3. Test in one-dimensional laminar flamelet

The ANNs are first tested on 1D laminar flamelet simulations. In this work, the training data set is the random data deriving from the 1D laminar flamelet simulations, however, the flamelet data were not used in the training process. Two different strain rates ($S = 50 \text{ s}^{-1}$ and 500 s^{-1}) are chosen to validate the ANNs. The mixture fraction space is discretised with 200 nodes and grid is refined around the stoichiometric mixture fraction. The ANNs simulated laminar flamelets is compared with the direct integration (DVODE) approach. In the both cases, the temperature profile, and all the major and minor species of the ANN prediction is in excellent agreement with the DI results, apart from the error of two minor species $\text{C}_2\text{H}_4\text{OH}$ and HOCH_2O in the case of $S = 50 \text{ s}^{-1}$. The mass fractions of both species are highly overpredicted by ANNs at very low mixture fraction locations ($z < 0.035$). While in the case of $S = 500 \text{ s}^{-1}$, the overprediction of HOCH_2O at low mixture is significantly reduced and results of $\text{C}_2\text{H}_4\text{OH}$ becomes very reasonable. The orders of magnitude of HOCH_2O and $\text{C}_2\text{H}_4\text{OH}$ is about 10^{-13} and 10^{-11} in the case of $S = 50 \text{ s}^{-1}$, reduced by a factor of 50 than $S = 500 \text{ s}^{-1}$, which are all much shorter than other species. Therefore, the ANN errors in the case of $S = 50 \text{ s}^{-1}$ are more obvious, due to the extremely small mass fractions of these two species.

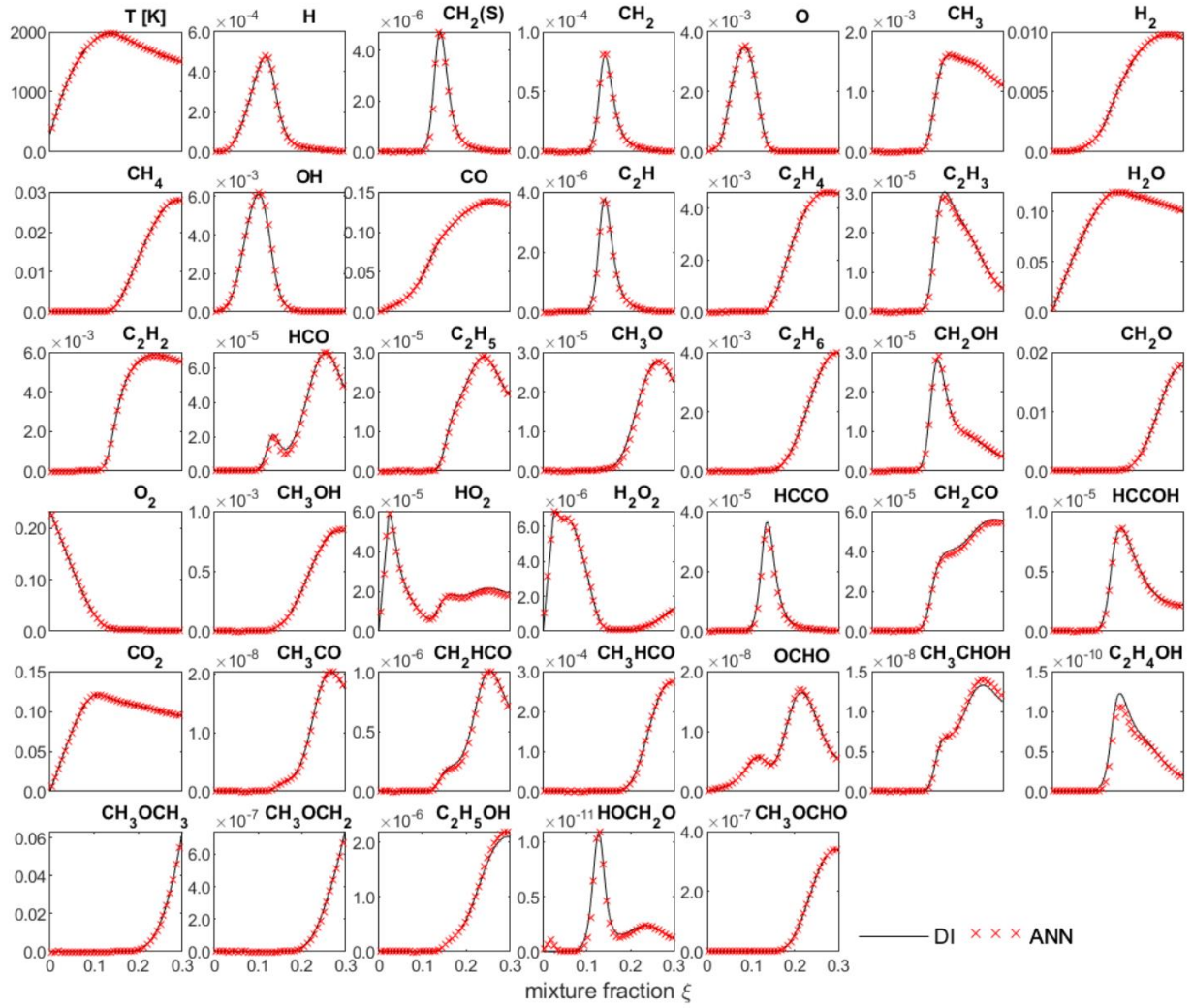
4. Test in turbulent diffusion flame - Sandia DME flame series

The Sandia piloted methane/air flame series are well-documented turbulent diffusion flames that have been extensively studied both experimentally and numerically [56, 57]. To explore the turbulence-flame interactions of more complex fuels, researchers have conducted experimental measurements [58–61] on a series of partially-premixed piloted DME/air jet flames with different inflow Reynolds numbers, known as the Sandia DME flame series. These flames have been chosen as benchmark flames on the TNF workshop (International Workshop on Measurement and Computation of Turbulent Nonpremixed Flames) for the turbulent combustion study of the oxygenated fuels. Hence, the Sandia DME flame series serves as an excellent validation platform for the HFRD-MMLP methodology, which aims to accelerate the real-time computation of DME fuel combustion.

The piloted Sandia DME flame D, developed by Coriton et al. [58], is analogous to the well-studied Sandia piloted methane/air flame D. It shares the identical stoichiometric mixture fraction, $\xi_{st} = 0.35$ and the bulk jet exit Reynolds



(a) strain rate $S = 50 \text{ s}^{-1}$



(b) strain rate $S = 500 \text{ s}^{-1}$

Figure 8: Simulated temperature and species mass fraction of the laminar flamelet using direct integration and ANNs

number $Re_D = 29,300$. In that study, Coriton et al. [58] reported velocity and mole fraction measurements of OH and CH_2O . LES-CMC simulations of Sandia DME flame D were conducted and compared to experimental measurements using the mechanisms developed by Zhao et al. [51] and Fischer et al. [50]. Fuest et al. [59] also measured the temperature and major species of Sandia DME flame D and F ($Re_F = 58,600$). Then, Popp et al. [62] performed an LES simulation of flame D using the flamelet/progress variable approach. Subsequently, Coriton et al. [60] reported the flow field and scalar measurements of the complete Sandia DME flame series, which includes flames with jet exit Reynolds numbers ranging from 29,300 (flame D) to 73,300 (flame G). Neuber et al. [61] later conducted LES-MMC simulations of the complete Sandia DME flame series using the detailed mechanism by Zhao et al. [51], and the comparison with the experiment showed good agreement. In our work, Sandia DME flame D and F are selected as a practical combustion problem for the trained ANNs.

	Unburnt gas composition						inflow operating condition			
	DME	C ₂ H ₂	H ₂	N ₂	O ₂	CO ₂	temperature [K]	bulk velocity [m/s]		Re _d
Jet	0.2	-	-	0.632	0.168	-	293	flame D	45.9	29,300
								flame F	91.8	58,600
Pilot	-	0.022	0.088	0.688	0.172	0.03	1900	flame D	1.1 (unburnt) 6.75 (burnt)	-
								flame F	2.1 (unburnt) 12.87(burnt)	
Co-flow	-	-	-	0.790	0.210	-	293	0.9		-

Table 2: Gas compositions and operating conditions of the turbulent DME/air flames

4.1. Simulation setup

A burner similar to the one used in the Sandia piloted methane/air flame series is employed for the Sandia DME flames. The main jet tube has an inner diameter of $D_{\text{jet}} = 7.45$ mm and is surrounded by an annular pilot with inner and outer diameters of 8 mm and 18.2 mm, respectively. The jet mixture consists of 80% air and 20% DME (volume fraction), resulting in a stoichiometric mixture fraction of 0.35. The bulk velocities of the main jet are 45.9 m/s and 91.8 cm/s for Sandia DME D and F flames, respectively. The velocity of the coflow is 0.9 m/s, and the operating temperature is 293 K. The pilot flow gas composition is equivalent to the product composition and temperature of a premixed DME/air flame at an equivalence ratio of 0.6. The bulk velocities of the burnt pilot are 6.74 cm/s and 12.87 cm/s for the DME D and F flames, respectively, with a burnt pilot gas temperature of 1900 K by the equilibrium simulation. Table 2 provides a summary of the unburnt gas compositions and operation conditions (bulk velocity and Reynolds number) of the main jet, pilot and co-flow for both flames.

The jet inflow velocity profile is pre-calculated using OpenFOAM DNS platform [63] in a fully developed pipeline flow. The diameter of the pipe matches the Sandia DME jet burner, and the length of the pipe is precisely 50 times of the jet diameter D_{jet} . The grid mesh consists of 1 million cells, employing an O grid at the pipe centre, and the boundary layer refinement with a maximum expansion ratio of 1.5. The DNS simulation is run for 5 convective flowthroughs, with a time step of 10^{-6} s.

The simulation domain is designed as a truncated cone, which gradually expands from a diameter of $12D_{\text{jet}}$ at the jet exit plane to $28D_{\text{jet}}$ at the height of $60D_{\text{jet}}$. A non-orthogonal O-grid is used for the meshing. The central region of the jet is resolved using a quasi-square grid with 36×36 cells. The outer quasi-annular area is meshed using a grid of 144 cells in the circumferential direction and 79 cells in the radial direction. The allocation of cells in the radial direction is as follows: 7 cells from the O-grid to the inner diameter of the jet, 4 cells on the jet rim thickness, 26 cells from the outer diameter of the jet towards pilot, and 42 cells from pilot towards the simulation domain boundary. The minimum cell size is approximately 0.07 mm at the jet rim and gradually expands towards the flame centre with a cell size of about 0.13 mm. The cell size also smoothly expands outwards in the radial direction with an expansion ratio of around 1.07. In the truncated cone-shape domain, the mesh size increases linearly as the diameter of the domain increases in the axial direction. The grid in the axial direction has 384 cells, with refinement near the inflow region.

Overall, the entire simulation domain consists of approximately 5.56 million cells.

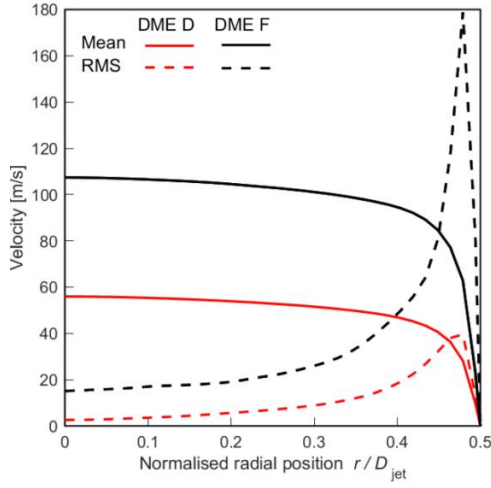


Figure 9: DNS simulation of the mean and RMS profiles of the jet inflow velocity for the Sandia DME flames using OpenFOAM

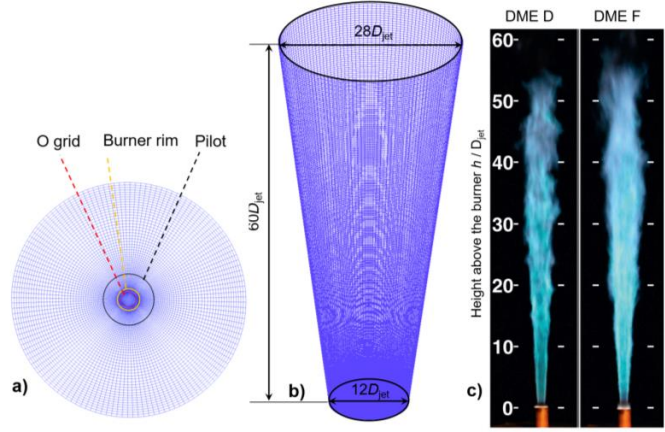


Figure 10: Grid of the computational domain and photographs of the Sandia DME flames and : (a) grid of the burner exit plane; (b) computational domain of the simulation; (c) photographs of the Sandia DME flame D and F. Image (c) from [60] with permission from Taylor & Francis ©

In this work, the simulations of the Sandia DME flames were performed with our in-house CFD program BOFFIN [64], where the LES-PDF method using the Eulerian stochastic approach [65] is implemented. Eight stochastic fields are used with the dynamic Smagorinsky model of Piomelli and Liu [66]. Equal scalar diffusivities are assumed with a sub-grid Schmidt number $\sigma_{sgs} = 0.7$. Numerical details about the BOFFIN codes can be referred in [64] and the thesis of Prasad [67]. The radiative heat loss is considered in our simulations using the optically thin radiation model and the radiative properties are based on the RADCAL model [68], in which the major species CH_4 , CO , CO_2 and H_2O are considered. Simulations of Sandia DME flame D and F are conducted in this work. For each flame, the direct integration (DVODE) and the HFRD-MMP method are used for computing the reaction source terms. All statistical quantities are collected for a period of more than 5 flow through times (based on the bulk jet velocity U_{jet}) once the flame becomes fully developed. In the LES-PDF simulation of the Sandia DME flame D, the flammability range by setting the mixture fraction range to $[0.05, 0.29]$ and a minimum reaction temperature of 800 K, which is inside the flammability range of the flamelet training samples.

4.2. Results for the ANNs simulations of Sandia DME Flame D

The radial mean and RMS profiles of temperature and concentrations of major and minor species for the flame D are presented in Figs. 11 and 12, respectively. These figures compare the simulation results obtained from the ANNs and DI methods at four different heights ($z/D_{jet} = 5, 10, 20$ and 40). Figure 11 illustrates the radial profiles of major species and temperature, with the ANN profiles represented by scattered dots and the DI profiles depicted using solid lines. Additionally, experimental data points are indicated by circles. The mean profiles are denoted by blue

dots/lines/circles, while the RMS profiles are presented by red dots/lines/circles. It is evident that ANN results exhibit excellent agreement with the DI results for temperature and most major species. Notably, the ANN results accurately predict both the peak magnitudes and their corresponding locations in the mean and RMS profiles.

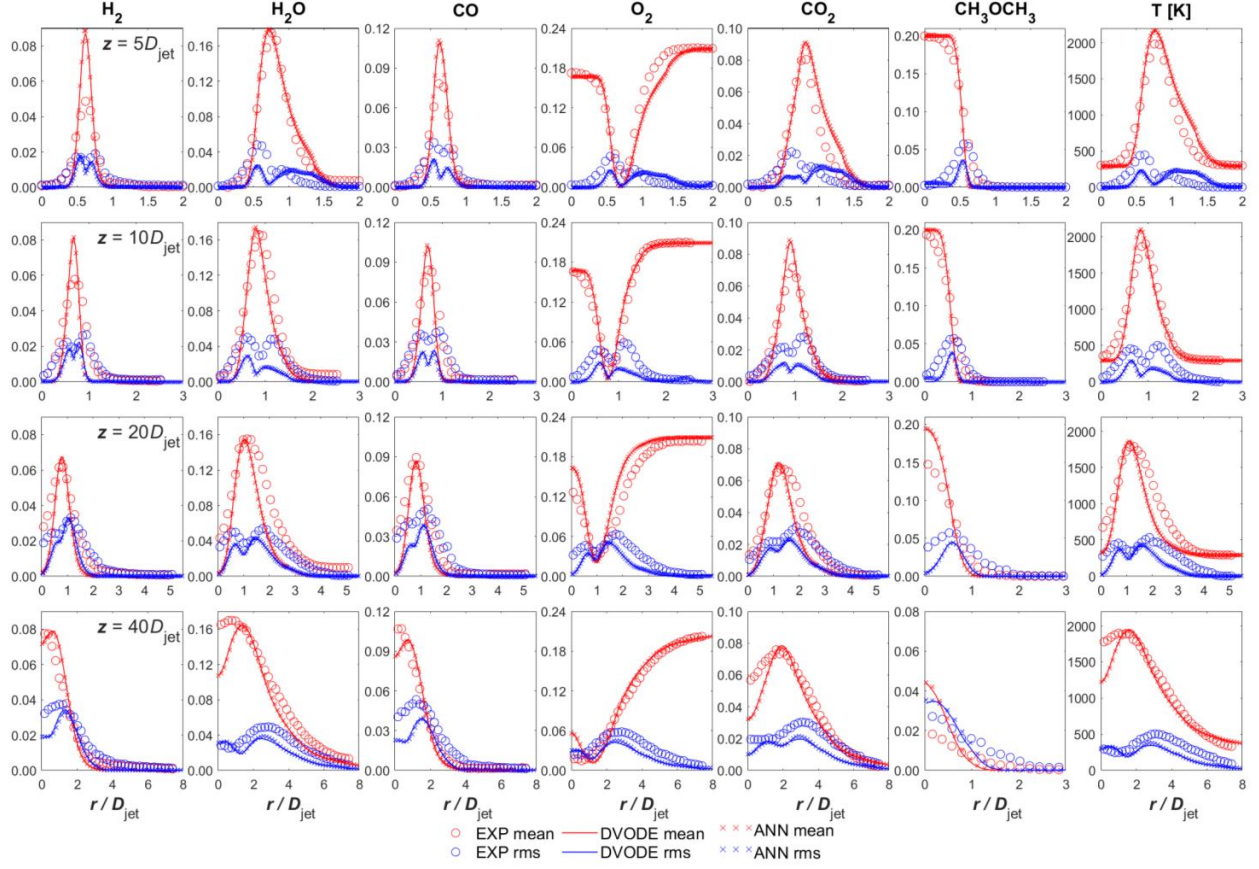


Figure 11: Radial profiles of temperature and mole fractions of major species for Sandia DME flame D, comparison among DI, ANNs and the experiment [59]

The simulated radial profiles of several minor species with mole fractions around 10^{-3} and 10^{-6} are displayed in Figs. 12a and 12b, respectively. The mean concentrations of all species predicted by the ANNs simulation show a quite good agreement with the results obtained from the DI method. However, there are some slight deviations in the predictions. Specifically, HO_2 and H_2O_2 are overpredicted by the ANNs by 5% - 10% at the low axial location ($z = 5D_{\text{jet}}$) while C_2H_6 and CH_3OH are overpredicted at the high axial location ($z = 40D_{\text{jet}}$). It is remarkable that the RMS profiles are almost exactly predicted by the ANNs when compared to the DI simulation. The only exception is the overprediction by 10% for species H, O, CH_2 and CH_2OH at the height of $z = 40D_{\text{jet}}$.

Figure 12c shows the simulation results of several minor species with very low mole fractions (less than 10^{-6}), which are typically not accounted for in simple mechanisms (*e.g.* such as GRI-1.2 and GRI-3.0 mechanisms [20, 49]). It is evident that the performance of the ANNs on these species is not as strong as those depicted in Figs. 11, 12a and 12b. For certain species, namely OCHO , CH_3OCH_2 and HOCH_2O , the peak values are obviously overpredicted

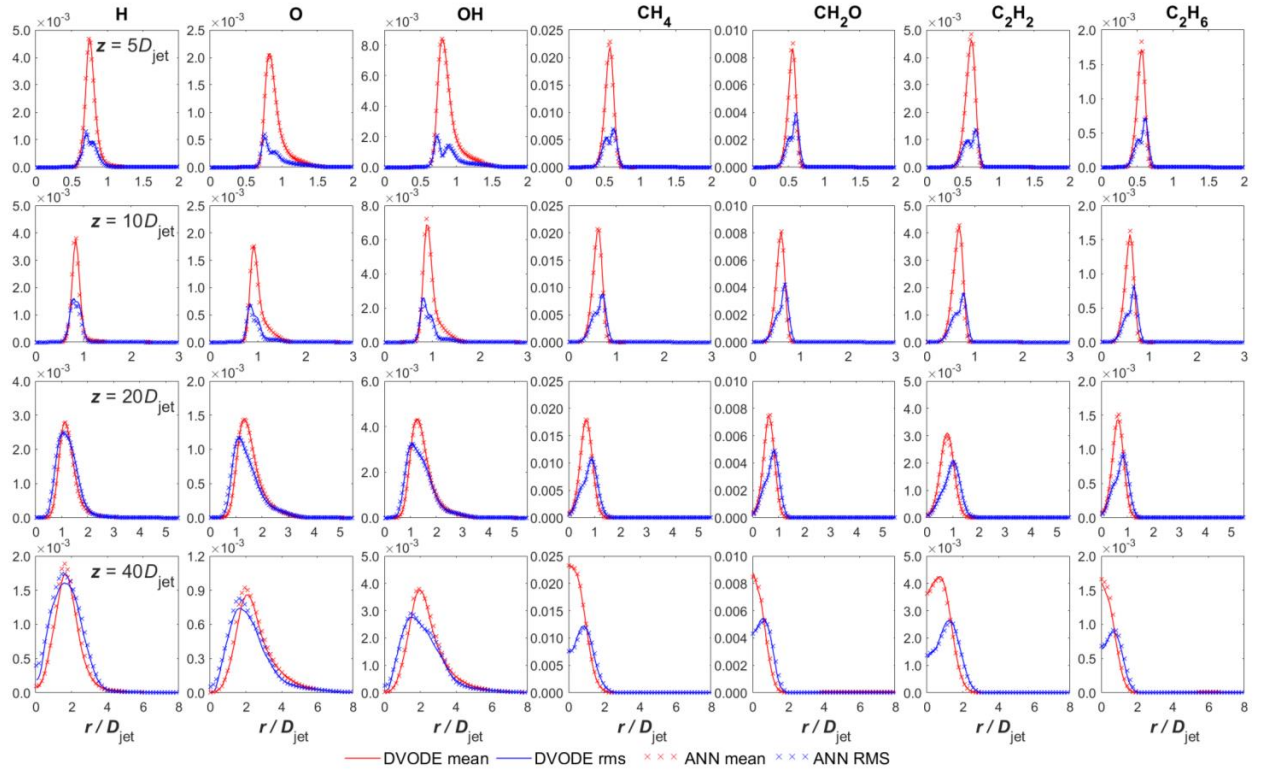
by a factor of 1.2 to 2 at the higher location $z = 40D_{\text{jet}}$. Conversely, the species $\text{C}_2\text{H}_5\text{OH}$ is underpredicted by approximately 20% - 30% within the height range of $z = 20D_{\text{jet}}$ to $40D_{\text{jet}}$. Accounting the considerably low mole fractions of these species, the ANN errors demonstrated here are deemed acceptable, unless one has specific interest in these species and necessitates simulation results of higher precision. Additionally, the peak locations of mean and RMS produced by ANNs still align well with those of DI, indicating the overall ANN profiles remain reasonable.

Figure 13 presents a comparison of contour plots illustrating the temperature field and mole fractions of two species, CO and CH_3O , simulated using both the ANNs and the DI approaches. Each plot consists of an instantaneous contour on the left and the mean contour on the right. Overall, the distributions of these scalar fields between the ANNs and DI show a high level of consistency. Examining the temperature field (Fig. 13a), the ANNs simulation accurately reproduces the absence of local extinction throughout the entire domain in the Sandia Flame D, as predicted by the DI simulation. The CO contour plot generated by the ANNs simulation captures the expansion of high CO values radially outwards from the mixing layer at a height of approximately $15D_{\text{jet}}$, where the turbulence starts to interact with the combustion reaction. The maximum peak of CO occurs at a height of $48D_{\text{jet}}$, indicating the presence of sufficient oxygen in the downstream combustion. A minor discrepancy between the ANNs and the DI results arises in the CH_3O mole fraction map, where DI predicts the disappearance of CH_3O at the height of $45D_{\text{jet}}$ while the ANNs results show its decay until a height of $50D_{\text{jet}}$. Nevertheless, both the ANNs and DI methods successfully captures the characteristics of the reactive scalar fields. Therefore, the HFRD-MMLP methodology demonstrates the ability to accurately reproduce the flame structure and species distribution.

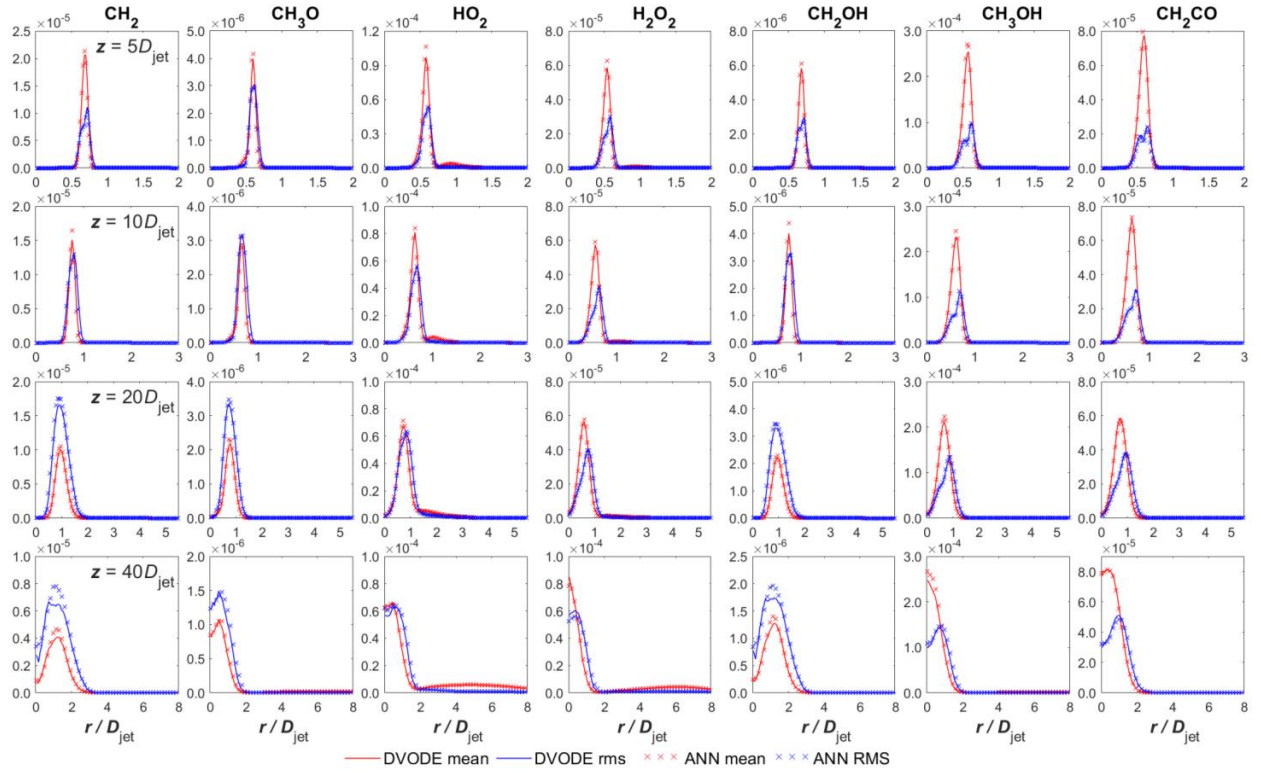
4.3. Results for the ANNs simulations of Sandia DME Flame F

The trained HFRD-MMLP ANNs are also used to predict the DME Sandia flame F, with a doubled jet Reynolds number than flame D. Figures 14 and 15 display the radial mean and RMS profiles of temperature and concentrations of major and minor species for flame F. A similar representation are used that the ANNs profiles are plotted using scattered dots while the DI profiles are illustrated by solid lines, calibrated with circles denoting experimental data. Similar conclusions as the DME flame D are drawn between the ANNs and DI predictions of the flame F:

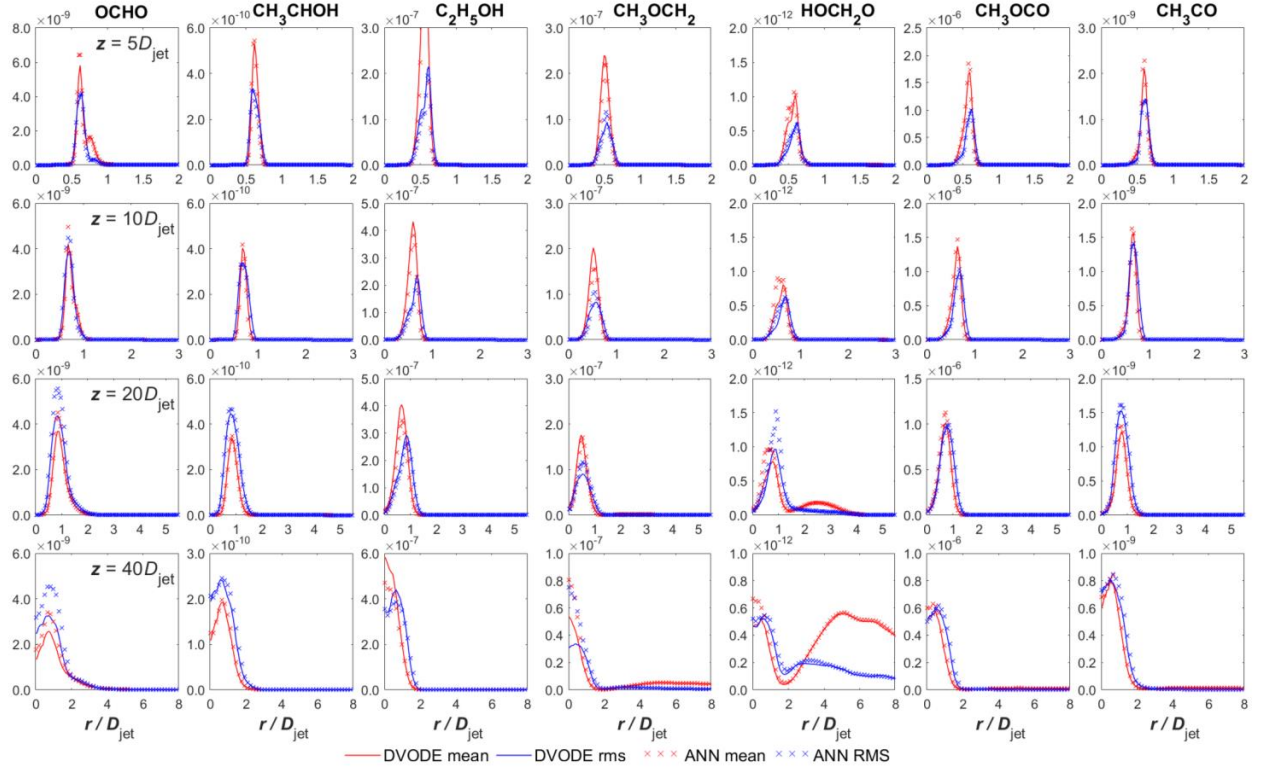
- (1) For the mean of temperature, major species (Fig. 14) and minor species with magnitudes of around 10^{-3} and 10^{-6} (Figs. 15a and 15b), the ANNs predictions align closely with the DI simulations. The notable discrepancies occur at a height of $40D_{\text{jet}}$, where DME (CH_3OCH_3), CH_4 , CH_2O , CH_3O , HO_2 and CH_3OH are overpredicted by a factor of 1.2-1.5, which relatively larger compared to the DME flame D. As the jet Reynolds number of Flame F is double than Flame D, stronger turbulence enhances the mixing of reactive scalars. Therefore, it is more likely to cause the concentrations of reactive scalars to exceed the composition bounds established by the training dataset and additional errors appears in predicting minor species.
- (2) For the minor species with smaller magnitudes (below 10^{-6} , Fig. 15c), the ANNs results show fine agreement with the DI simulation in the upstream regions. In the ANNs simulation at the height of $40D_{\text{jet}}$, species CH_3OCH_2 and CH_3OCHO are overpredicted by approximately a factor of 1.5-1.8, and HOCH_2O and CH_3OCO are overpredicted



(a) part 1, mole fraction of around 10^{-3}



(b) part 2, species mole fraction of around 10^{-6}



(c) part 3, species mole fraction below 10^{-6}

Figure 12: Radial profiles of mole fractions of minor species for the flame D

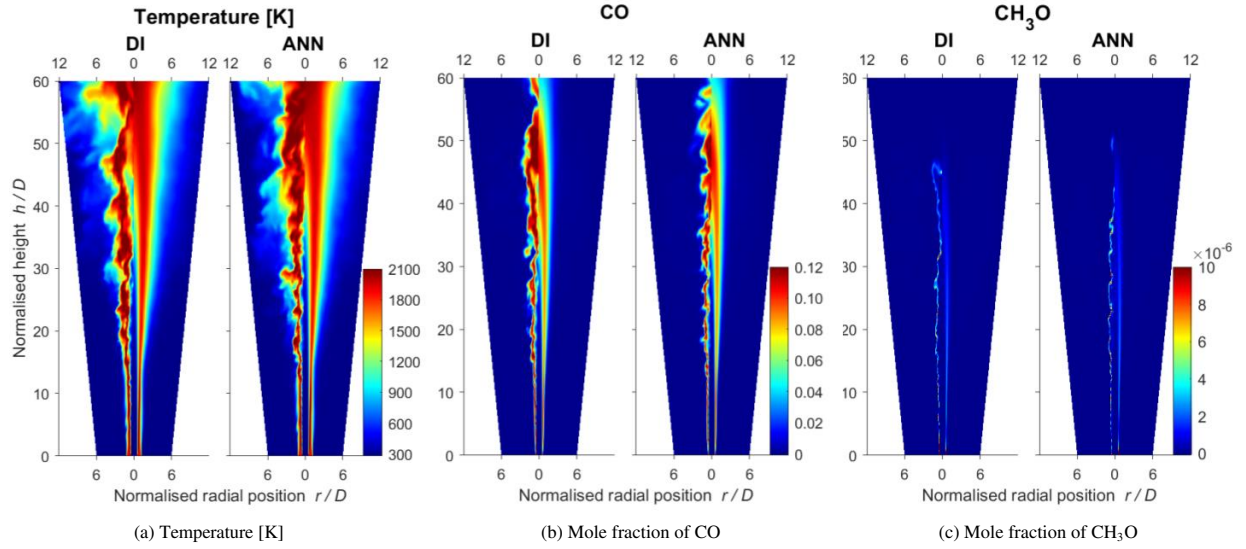


Figure 13: Comparison of contour plots of reactive scalar fields of Sandia DME flame D between the ANNs and DI simulations (instantaneous and mean contours are combined at the left and right part of each plot)

390 by approximately a factor of 1.2. In contrast, CH₃CHOH is underpredicted by 20% in the ANN simulation compared
 391 to the DI results

(3) The RMS of the temperature profile and all species are basically consistent between the ANNs and DI simulations. Some overpredictions for minor species can be found at the height of $40D_{\text{jet}}$, *i.e.* CH_2 , CH_3O , OCHO , CH_3CHOH , CH_3OCH_2 and CH_3OCO .

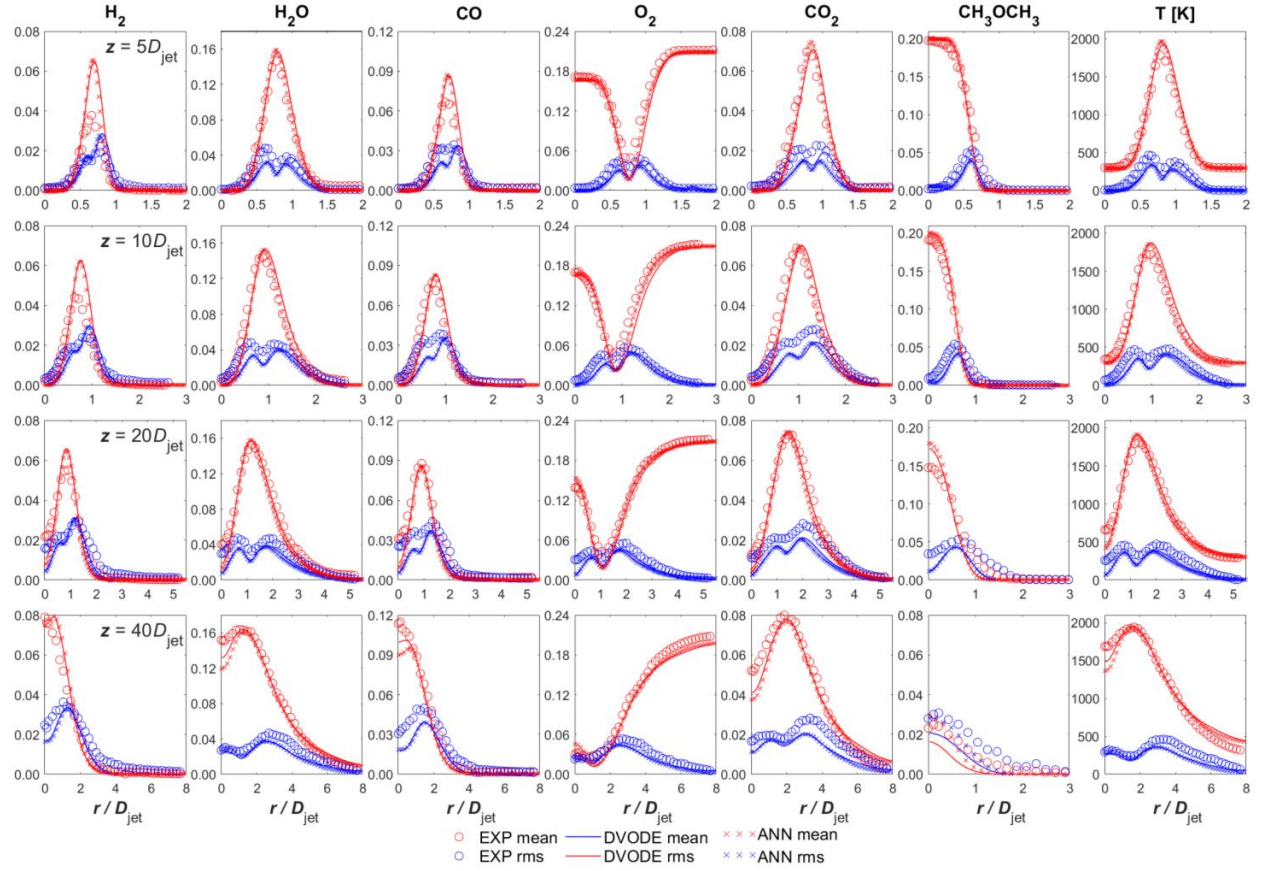
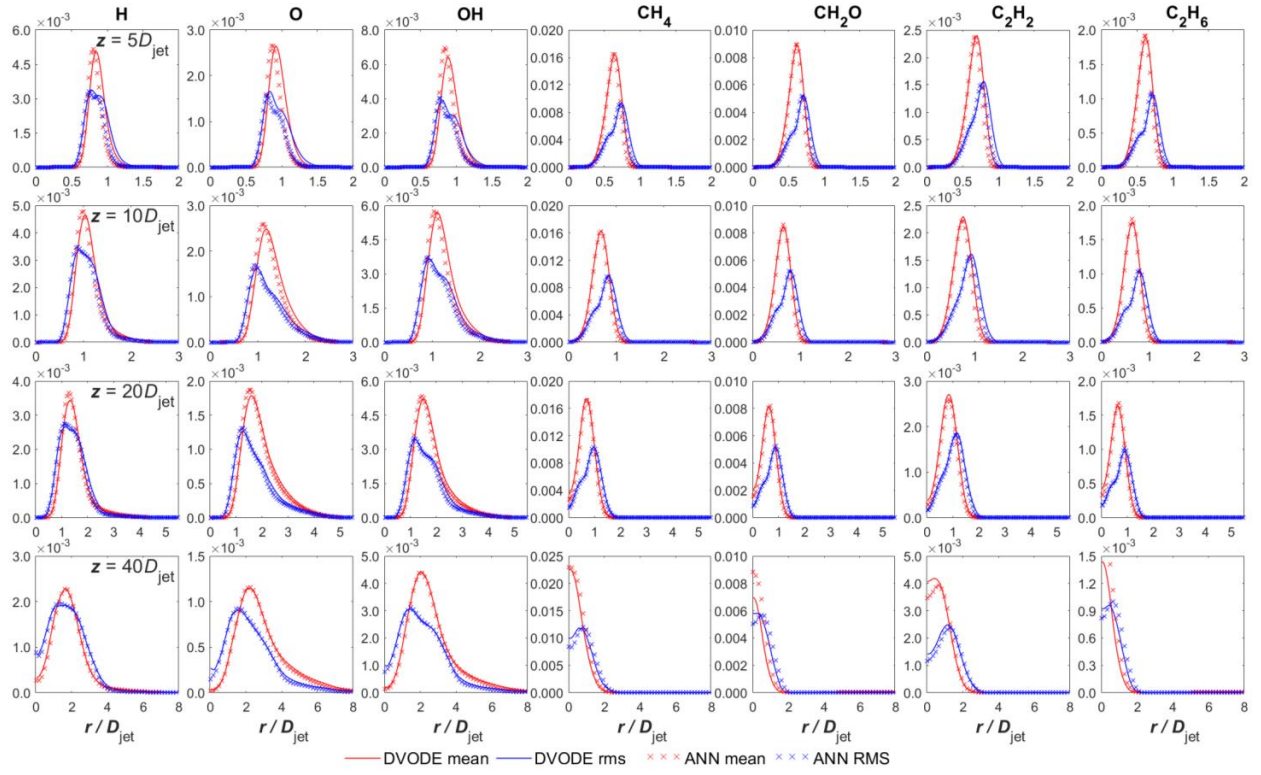


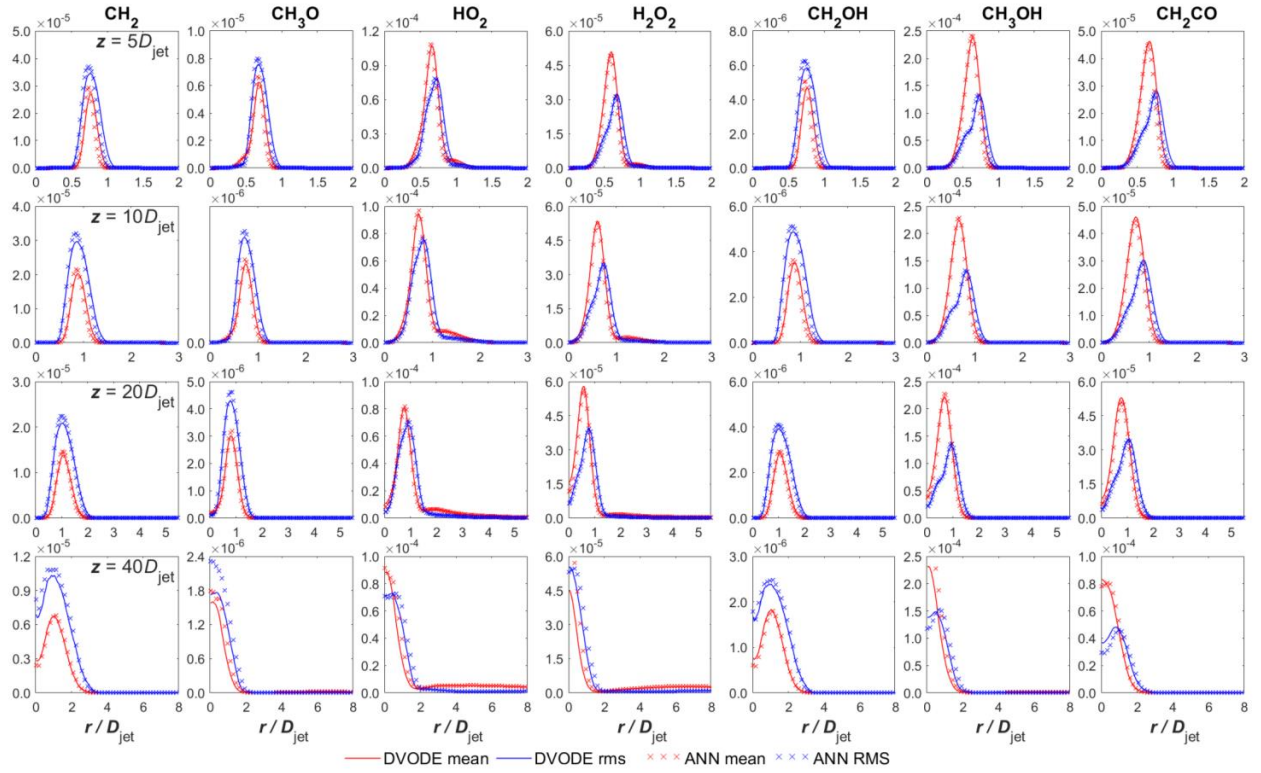
Figure 14: Radial profiles of temperature and mole fractions of major species for Sandia DME flame F, comparing among DI, ANNs and the experiment [59]

4.4. Element conservation check

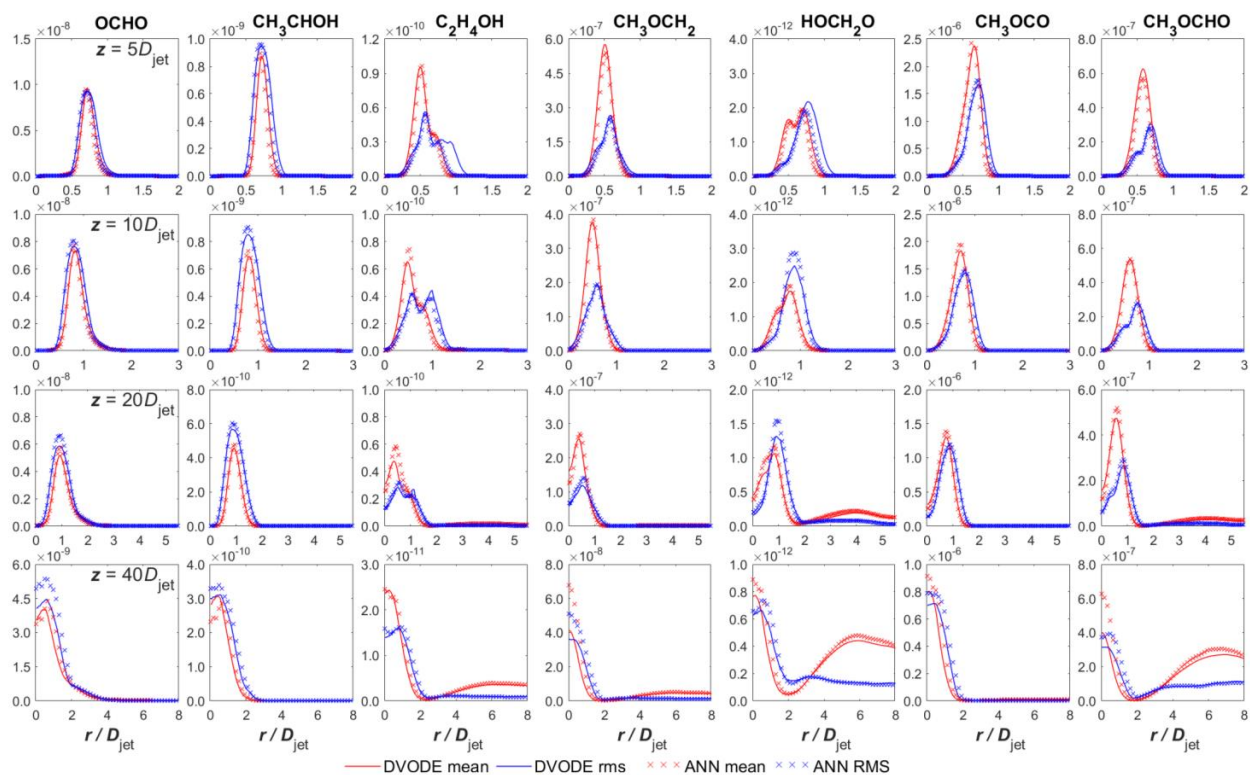
In fact, the element conservation constraint is not implemented in the HFRD-MMLP method. However, two means implemented in this study indirectly help to improve the accuracy of the ANNs on the element conservation. First of all, in the process of data generation, the element constraints have been incorporated to ensure generate physically realistic data (see Table 1), with the H/C ratio between 2.9 and 3.1 and O/N ratio between 0.255 and 0.275. Second, the sum mass fraction of all species will be normalised after the chemistry calculation by ANNs. With these two means, the ANNs achieve high prediction accuracy in the laminar flamelets (see Section 3) and the Sandia DME turbulent flames (Sec 4). Here, we calculate the error of element conservation using ANNs after 1000 time steps. The data samples are from the LES-PDF simulation of the Sandia DME D flames using the DVODE approach. The results show that the average error for all elements by ANNs calculation are below 0.25% over 1000 time steps ($1 \times 10^{-3}\text{s}$).



(a) part 1, mole fraction of around 10^{-3}



(b) part 2, species mole fraction of around 10^{-6}



(c) part 3, species mole fraction below 10^{-6}

Figure 15: Radial profiles of mole fractions of minor species for the flame F

405 That means the ANNs prediction is of high accuracy and can ensure the element conservation.

Elements	C	H	O	N
Average relative errors	0.16%	0.22%	0.09%	0.07%
Percentage of samples with the relative error above 1.0%	0.27%	0.29%	0.01%	0.00%

Table 3: Relative errors for all elements after 1000 time steps using ANNs with over 2 million data samples collected from the LES-PDF simulations of Sandia DME flames

406 4.5. CPU time acceleration in the ANNs simulations

407 The average CPU time comparison of the Sandia DME series flame simulations are presented in Table 4. When
 408 compared to DI, the ANN approaches demonstrates a significant reduction in the time required for the reaction step,
 409 with a decrease of 95.7%. This reduction corresponds to a remarkable speed-up ratio of 16.2, leading to a speed-up
 410 ratio of 10.0 in the total computation time. Furthermore, the percentage of reaction time to total time decreases from
 411 95.7% to 58.7%, indicating a substantial reduction in the computation burden associated with the chemical kinetics
 412 calculation.

413 Now, let us compare the CPU performance between the current study, utilising the HFRD-MMLP thermochem-
 414 istry tabulation method for the DME mechanism, with the previous work, employing the same approach for GRI-1.2

Mechanisms Applications		DME Sandia DME flames(this work)			GRI-1.2 Sydney CH ₄ /H ₂ HM1 flames		
Basic Information	species / reactions	55 / 290			32 / 177		
	CPU cores	448			200		
	Total cells / Reacting (%)	5,560,000 / 35.1%			2,860,000 / 21.0%		
time cost (normalised)		DVODE	ANNs	speed-up	DVODE	ANNs	speed-up
	Reaction time t_R	220.9 s (1)	13.59 s (0.062)	16.2	70.82 s (1)	5.17 s (0.073)	10.0
	transport time	LES	2.72 s	2.61 s	–	13.75 s (0.19)	13.87 s (0.20)
		PDF	7.32 s	7.04 s	–		
	Total time t_T	230.9 s (1.045)	23.24 s (0.105)	13.7	84.57 s (1.19)	19.04 s (0.27)	4.4
	t_R/t_T	95.7%	58.7%	–	83.7%	27.2%	–

Table 4: Average CPU time for the Sandia DME flame series simulations

mechanism [49], consisting of only 30 species. The ANNs with GRI-1.2 mechanism was applied in turbulent non-premixed Sydney CH₄/H₂ HM1 flames. The speed-up effect achieved by the ANNs for the DME mechanism is more significant compared to the GRI-1.2 ANNs, in terms of reaction time and total time costs. In particular, the speed-up ratio for the total computation time in the DME-ANNs simulation is over double than that for the GRI-1.2 ANNs. Therefore, the HFRD-MMLP method are more suitable and effective for large-size mechanisms, such as the DME mechanism, where it demonstrates significant speed-up benefits in terms of both reaction time and overall computation time.

5. Conclusions

In this study, the HFRD (hybrid flamelet/random data) - MMLP (multiple multilayer perceptrons) thermochemistry tabulation methodology is employed to accelerate the real-time computation of a large-size mechanism, the DME mechanism [51] for pyrolysis and combustion. The HFRD method incorporates element constraints that ensure the generation of physically realistic data, specifically for the oxygenated fuel. Additionally, the MMLP approach is employed to improve the ANNs prediction accuracy, with subsequent MLP being dedicated to a different output range. In total, 132 MLPs are trained to represent the DME mechanism.

The ANNs are initially tested on 1-D laminar flamelet simulations under different strain rates, yielding excellent results for flamelets with varying strain rates. For flamelet simulations with low strain rates, the ANN results of most species are accurate, apart from some errors observed for a few minor species of very low mass fractions. Subsequently, the ANN methodology is further validated through the simulation of turbulent flames, specifically Sandia DME D and F flames. The simulation results are compared with those obtained using the direct integration method and show overall very good agreement for the temperature, major species and the majority of minor species. Although there are some minor species that exhibit overprediction at the downstream location, the peak magnitude locations are still accurately predicted by the ANNs. In general, the ANN simulation results for the Sandia flame series demonstrate good agreement with the DI simulation results.

The HFRD-MMLP achieves a significant speed-up factor of over 16 and 10 for the reaction source term and total

computation time, respectively, in the LES-PDF simulation using the DME mechanism. These results provide strong evidence that the HRFD-MMLP method in fact is highly effective and capable of achieving a substantial speed-up ratio for large-size mechanisms, thus reducing the need for computationally expensive integrations of highly stiff ODEs.

Acknowledgements

Funding for the research sponsored by the National Natural Science Foundation of China under Grant No. 51925603 and Shanghai Pujiang Program under Grant No. 22PJ142800 is gratefully acknowledged.

References

- [1] C. Arcoumanis, C. Bae, R. Crookes, E. Kinoshita, The potential of di-methyl ether (DME) as an alternative fuel for compression-ignition engines: A review, *Fuel* 87 (2008) 1014–1030.
- [2] S. Park, C. Lee, Combustion performance and emission reduction characteristics of automotive DME engine system, *Progress in energy and combustion science* 39 (2013) 147–168.
- [3] G. Thomas, B. Feng, A. Veeraragavan, M. Cleary, N. Drinnan, Emissions from DME combustion in diesel engines and their implications on meeting future emission norms: A review, *Fuel processing technology* 119 (2014) 286–304.
- [4] N. Peters, *Turbulent Combustion*, Cambridge University Press, 2000.
- [5] D. Veynante, L. Vervisch, Turbulent combustion modeling, *Progress in Energy and Combustion Science* 28 (2002) 193–266.
- [6] T. Poinso, D. Veynante, *Theoretical and Numerical Combustion*, 2nd ed., Edwards, 2005.
- [7] A. Shakeri, K. Mazaheri, M. Owliya, Using sensitivity analysis and gradual evaluation of ignition delay error to produce accurate low-cost skeletal mechanisms for oxidation of hydrocarbon fuels under high-temperature conditions, *Energy Fuels* 31 (2017) 11234–11252.
- [8] Q. Wang, Skeletal mechanism generation for high-temperature combustion of $H_2/CO/C_1-C_4$ hydrocarbons, *Energy & Fuels* 27 (2013) 4021–4030.
- [9] T. Bolshova, V. Shvartsberg, A. Dmitriev, D. Knyazkov, Flame structure and a compact reaction mechanism for combustion of dimethyl ether at atmospheric pressure, *Fuel* 255 (2019) 115752.
- [10] W. Sun, Z. Chen, X. Gou, Y. Ju, A path flux analysis method for the reduction of detailed chemical kinetic mechanisms, *Combustion and Flame* 157 (2010) 1298–1307.
- [11] W. Wang, X. Gou, An improved path flux analysis with multi generations method for mechanism reduction, *Combustion Theory and Modelling* 20 (2016) 203–220.
- [12] S. Lam, D. Goussis, The CSR method for simplifying kinetics, *International Journal of Chemical kinetics* 26 (1994) 461–486.
- [13] P. Konaviitis, S. Rigopoulos, W. Jones, A methodology for derivation of RCCE-reduced mechanisms via CSP, *Combustion and Flame* 183 (2017) 126–143.
- [14] P. Konaviitis, S. Rigopoulos, W. Jones, Reduction of a detailed chemical mechanism for a kerosene surrogate via RCCE-CSP, *Combustion and Flame* 194 (2018) 85–106.
- [15] X. Gao, S. Yang, W. Sun, A global pathway selection algorithm for the reduction of detailed chemical kinetic mechanisms, *Combustion and Flame* 167 (2016) 238–247.
- [16] J. Cheng, C. Zou, Q. Lin, High-temperature oxidation of methyl isopropyl ketone: a shock tube experiment and a kinetic model, *Combustion and Flame* 209 (2019) 376–388.
- [17] A. Paykani, C. Frouzakis, K. Boulouchos, Numerical optimization of methane-based fuel blends under engine-relevant conditions using a multi-objective genetic algorithm, *Applied Energy* 242 (2019) 1712–1724.

- [18] M. Lu, Z. Fu, X. Yuan, G. Sun, G. Jia, Study of the reduced kinetic mechanism of methane/dimethyl ether combustion, *Fuel* 303 (2021) 121308.
- [19] Y. Ding, W. Zhang, L. Yu, K. Lu, The accuracy and efficiency of GA and PSO optimization schemes on estimating reaction kinetic parameters of biomass pyrolysis, *Energy* 176 (2019) 582–588.
- [20] G. Smith, D. Golden, M. Frenklach, N. Moriarty, B. Eiteneer, M. Goldenberg, C. Bowman, R. Hanson, S. Song, W. Gardiner Jr., V. Lissianski, Z. Qin, GRI-3.0 Mechanism, 1995. URL: http://www.me.berkeley.edu/gri_mech/.
- [21] J. Appel, H. Bockhorn, M. Frenklach, Kinetic modeling of soot formation with detailed chemistry and physics: laminar premixed flames of C2 hydrocarbons, *Combustion and Flame* 121 (2000) 122–136.
- [22] G. Blanquart, P. Pepiot-Desjardins, H. Pitsch, Chemical mechanism for high temperature combustion of engine relevant fuels with emphasis on soot precursors, *Combustion and Flame* 156 (2009) 588–607.
- [23] Y. Wang, A. Raj, S. Chung, A PAH growth mechanism and synergistic effect on PAH formation in counterflow diffusion flames, *Combustion and Flame* 160 (2013) 1667–1676.
- [24] A. Briones, S. Som, S. Aggarwal, Effect of multistage combustion on nox emissions in methane–air flames, *Combustion and Flame* 149 (2007) 448–462.
- [25] A. Liu, C. Garcia, F. Sewerin, B. William, S. Rigopoulos, Population balance modelling and laser diagnostic validation of soot particle evolution in laminar ethylene diffusion flames, *Combustion and Flame* 221 (2020) 384–400.
- [26] B. Sun, S. Rigopoulos, A. Liu, Modelling of soot formation and aggregation in turbulent flows with the LES-PBE-PDF approach and a conservative sectional method, *Combustion and Flame* 242 (2022) 112152.
- [27] J.-Y. Chen, W. Kollmann, R. Dibble, PDF modeling of turbulent nonpremixed methane jet flames, *Combustion Theory and Modelling* 64 (1989) 315–346.
- [28] U. Mass, S. Pope, Implementation of simplified chemical kinetics based on intrinsic low-dimensional manifolds, *Symposium (International) on Combustion* 24 (1992) 1231–1250.
- [29] S. Pope, Computationally efficient implementation of combustion chemistry using in situ adaptive tabulation, *Combustion Theory and Modelling* 1 (1997) 41–63.
- [30] F. Christo, A. Masri, E. Nebot, T. Turanyi, Utilizing artificial neural network and repro-modelling in turbulent combustion, in: *IEEE International Conference on Neural Networks - Conference Proceedings*, 2, 1995, pp. 911–916.
- [31] F. Christo, A. Masri, N. E.M., Artificial neural network implementation of chemistry with PDF simulation of H2/CO2 flames, *Combustion and Flame* 106 (1996) 406–427.
- [32] J. Blasco, N. Fuego, C. Dopazo, J. Ballester, Modelling the temporal evolution of a reduced combustion chemical system with an artificial neural network, *Combustion and Flame* 113 (1998) 38–52.
- [33] J. Blasco, N. Fuego, J. Larroya, C. Dopazo, Y.-J. Chen, A single-step time-integrator of a methane–air chemical system using artificial neural networks, *Computers & Chemical Engineering* 23 (1999) 1127–1133.
- [34] J.-Y. Chen, J. Blasco, N. Fuego, C. Dopazo, An economical strategy for storage of chemical kinetics: Fitting *in situ* adaptive tabulation with artificial neural networks, *Proceedings of the Combustion Institute* 28 (2000) 115–121.
- [35] J. Blasco, N. Fuego, C. Dopazo, Y.-J. Chen, A self-organizing-map approach to chemistry representation in combustion applications, *Combustion Theory and Modelling* 4 (2000) 61–76.
- [36] A. Chatzopoulos, S. Rigopoulos, A chemistry tabulation approach via rate controlled constrained equilibrium (RCCE) and artificial neural networks (ANNs), with application to turbulent non-premixed CH4/H2/N2 flames, *Proceedings of the Combustion Institute* 185 (2013) 1465–1473.
- [37] L. Franke, A. Chatzopoulos, S. Rigopoulos, Tabulation of combustion chemistry via artificial neural networks (ANNs): Methodology and application to LES-PDF simulation of sydney flame L, *Combustion and Flame* 185 (2017) 245–260.
- [38] V. Ihme, C. Schmitt, H. Pitsch, Optimal artificial neural networks and tabulation methods for chemistry representation in les of a bluff-body swirl-stabilized flame, *Proceedings of the Combustion Institute* 32 (2009) 1527–1535.

- [39] T. Readshaw, T. Ding, S. Rigopoulos, W. Jones, Modeling of turbulent flames with the large eddy simulation–probability density function (LES–PDF) approach, stochastic fields, and artificial neural networks, *Physics of Fluids* 33 (2021) 035154.
- [40] K. Wan, C. Barnaud, L. Vervisch, P. Domingo, Chemistry reduction using machine learning trained from non-premixed micro-mixing modeling: Application to dns of a syngas turbulent oxy-flame with side-wall effects, *Combustion and Flame* 220 (2020) 119–129.
- [41] M. Hansinger, Y. Ge, M. Pflitzner, Deep residual networks for flamelet/progress variable tabulation with application to a piloted flame with inhomogeneous inlet, *Combustion Science and Technology* 194 (2022) 1587–1613.
- [42] A. Kempf, F. Flemming, J. Janicka, Investigation of lengthscales, scalar dissipation, and flame orientation in a piloted diffusion flame by LES, *Proceedings of the Combustion Institute* 30 (2005) 557–565.
- [43] B. Sen, S. Menon, Turbulent premixed flame modeling using artificial neural networks based chemical kinetics, *Proceedings of the Combustion Institute* 32 (2009) 1605–1611.
- [44] B. Sen, S. Menon, Linear eddy mixing based tabulation and artificial neural networks for large eddy simulations of turbulent flames, *Combustion and Flame* 157 (2010) 62–74.
- [45] B. Sen, E. Hawkes, S. Menon, Large eddy simulation of extinction and reignition with artificial neural networks based chemical kinetics, *Combustion and Flame* 157 (2010) 566–578.
- [46] J. An, G. He, K. Luo, F. Qin, B. Liu, Artificial neural network based chemical mechanisms for computationally efficient modeling of hydrogen/carbon monoxide/kerosene combustion, *International Journal of Hydrogen Energy* 45 (2020) 159–190.
- [47] T. Ding, T. Readshaw, S. Rigopoulos, W. Jones, Machine learning tabulation of thermochemistry in turbulent combustion: An approach based on hybrid flamelet/random data and multiple multilayer perceptrons, *Combustion and Flame* 231 (2021) 111493.
- [48] T. Ding, S. Rigopoulos, W. Jones, Machine learning tabulation of thermochemistry of fuel blends, *Application in Energy and Combustion Science* 12 (2022) 100086.
- [49] M. Frenklach, H. Wang, M. Goldenberg, G. Smith, D. Golden, C. Bowman, R. Hanson, W. Gardiner, V. Lissianski, GRI-mech - an optimized detailed chemical reaction mechanism for methane combustion, Technical Report GRI-95/0058, Gas Research Institute, 1995.
- [50] S. Fischer, F. Dryer, C. H.J., The reaction kinetics of dimethyl ether. I: High-temperature pyrolysis and oxidation in flow reactors, *International Journal of Chemical Kinetics* 32 (2000) 713–740.
- [51] Z. Zhao, M. Chaos, A. Kazakov, F. Dryer, The reaction kinetics of dimethyl ether. I: High-temperature pyrolysis and oxidation in flow reactors, *International Journal of Chemical Kinetics* 40 (2008) 1–18.
- [52] U. Burke, K. Somers, P. O’Toole, C. Zinner, N. Marquet, G. Bourque, E. Petersen, W. Metcalfe, Z. Serinyel, C. H.J., An ignition delay and kinetic modeling study of methane, dimethyl ether, and their mixtures at high pressures, *Combustion and Flame* 162 (2015) 315–330.
- [53] H. Demuth, M. Beale, O. De Jess, M. Hagan, Neural network design, Martin Hagan, 2014.
- [54] D. Marquardt, An algorithm for least squares estimation of nonlinear parameters, *SIAM Journal on Applied Mathematics* 11 (1987) 431–441.
- [55] P. Brown, G. Byrne, A. Hindmarsh, VODE: a variable-coefficient ODE solver, *SIAM Journal on Scientific and Statistical Computing* 10 (1989) 1038–1051.
- [56] C. Schneider, A. Dreizler, J. Janicka, E. Hassel, Flow field measurements of stable and locally extinguishing hydrocarbon-fuelled jet flames, *Combustion and Flame* 135 (2003) 185–190.
- [57] R. Barlow, J. Frank, Effects of turbulence on species mass fractions in methane/air jet flames, *Symposium (International) on Combustion* 135 (1998) 185–190.
- [58] B. Coriton, M. Zendejdel, S. Ukai, A. Kronenburg, O. Stein, S.-K. Im, M. Gamba, J. Frank, Imaging measurements and LES-CMC modeling of a partially-premixed turbulent dimethyl ether/air jet flame, *Proceedings of the Combustion Institute* 35 (2015) 1251–1258.
- [59] F. Fuest, G. Magnotti, R. Barlow, J. Sutton, Scalar structure of turbulent partially premixed dimethyl ether/air jet flames, *Proceedings of the Combustion Institute* 35 (2015) 1235–1242.
- [60] B. Coriton, S. Im, M. Gamba, J. Frank, Flow field and scalar measurements in a series of turbulent partially-premixed dimethyl ether/air jet flames, *Combustion and Flame* 180 (2017) 40–52.
- [61] G. Neuber, F. Fuest, J. Kirchmann, A. Kronenburg, O. Stein, S. Galindo-Lopez, M. Cleary, R. Barlow, B. Coriton, J. Frank, J. Sutton,

563 Sparse-lagrangian MMC modelling of the sandia DME flame series, *Combustion and Flame* 208 (2019) 110–121.

564 [62] S. Popp, F. Hunger, S. Hartl, D. Messig, B. Coriton, J. Frank, F. Fuest, C. Hasse, LES flamelet-progress variable modeling and measurements
565 of a turbulent partially-premixed dimethyl ether jet flame, *Combustion and Flame* 162 (2015) 3016–3029.

566 [63] OpenFoam User Guide, 2021.

567 [64] W. Jones, F. di Mare, A. Marquis, LES-BOFFIN: user’s guide, 2002.

568 [65] W. Jones, V. Prasad, Large eddy simulation of the sandia flame series (D-F) using the Eulerian stochastic field method, *Combustion and*
569 *Flame* 157 (2010) 1621–1636.

570 [66] U. Piomelli, J. Liu, Large-eddy simulation of rotating channel flows using a localized dynamic model, *Physics of Fluids* 7 (1995) 839–848.

571 [67] V. Prasad, Large eddy simulation of partially premixed turbulent combustion, Ph.D. thesis, Imperial College London, 2011.

572 [68] W. Grosshandler, Radcal: a narrow-band model for radiation, calculations in a combustion environment, in: NIST technical note: 1402,
573 1993.