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ORIGINAL PAPER

NO formation analysis of turbulent non-premixed coaxial methane/air diffusion flame

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Abstract Natural gas combustion is one of the primary sources of harvesting energy for various processes and has gained a wide attention during the past decade. One of the most recent applications of natural gas combustion can be found in non-premixed combustion of methane in a coflow burner system. One of the main environmental concerns that arises from the natural gas combustion is the formation of NO produced by thermal NO and prompt NO mechanisms. Current paper is devoted on an examination of a 2D numerical simulation of turbulent non-premixed coaxial methane combustion in air enclosed by an axisymmetric cylindrical chamber to study the effects of species concentrations of reactants on NO formation, their individual contributions, and the chamber outlet temperature. A finite-volume staggered grid method is utilized to solve conservation equations of mass, energy, momentum, and species concentrations. In order to handle radiation heat transfer, discrete transfer method is used to solve radiation equation. Utilizing weighted-sum-of-gray-gases model, based on the newly obtained high-temperature molecular spectroscopic data, local variations of species absorption coefficients are taken into account. To calculate NO concentration, a single- or joint-variable probability density function in terms of a normalized temperature, mass fractions of species, or a combination of both is employed. Plus, published relevant experimental data are used to validate temperature and species concentration fields. It is shown that a decrease in N_2 concentration contributes to reducing NO. More importantly for higher O_2 mass fraction, thermal NO formation becomes the dominant mechanism responsible for NO emission.

Keywords Turbulent flame · NO environmental pollution · Natural gas combustion · Finite-volume method · High-temperature molecular spectroscopic database

Introduction

The nitrogen oxides formed in combustion systems are a main source of environmental pollution. Nitrogen oxides consist of nitric oxide (NO), nitrogen dioxide (NO_2), and nitrous oxide (N_2O). NO and NO_2 are collectively referred to NO. At the molecular level within turbulent flames, the NO formation can be related to three distinct chemical kinetic processes. These three processes responsible for NO formation are thermal NO, prompt NO, and fuel NO. Thermal NO is formed through high-temperature oxidation of the diatomic nitrogen found in combustion chamber system. Prompt NO is produced by the reaction of atmospheric nitrogen, N_2, with radicals such as C, CH, and CH_2 fragments derived from fuel, and finally fuel NO is produced by oxidation of nitrogen contained in the fuel.

Nowadays, numerical simulations absorb attention because of their accuracy and economical cost in resembling the actual phenomena (Al-sarraf et al. 2015; Liang et al. 2015; Poozesh et al. 2015; Jebali et al. 2015). It is also proved to be an inexpensive but efficient tool in simulating combustion process (Jeshvaghani et al. 2014; Yan et al. 2014; Stroh et al. 2015). Post-combustion cleanup of NO is feasible, but simulation of the combustion process can control NO economically. Therefore, the numerical simulation of NO formation in turbulent combustion is frequently studied in the optimization design of

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low-NO burners and furnaces. The NO formation process for turbulent combustion involves turbulent simulation, combustion simulation, and NO formation simulation. So comprehensive modeling of NO reaction processes in combustion systems requires simulation of both the turbulent fluid dynamics and the chemical kinetics in the modeled system. As some of the recent numerical investigations on NO formation in a turbulent combustion process, Zhou et al. (2002) used unified second-order moment (USM) model to investigate the formation of NO in turbulent combustion processes by solving conservation equations including mass, momentum, energy, and chemical species equations. In order to handle mean flow characteristics for turbulent flow, standard $k - \varepsilon$ turbulence model was used. A one-step chemical reaction which includes only two chemical species of O_2 and fuel was assumed. Considering the complexity and variety of the involved reactions, this assumption might bear further reconsideration. Their study was devoted to analyzing the diffusion of CH_4 in the free jet air to predict the NO formation. At the end, they concluded that USM model is capable of simulating the interaction of turbulence with detailed chemical kinetics, whereas it is much economical than other refined models. Hence, it can be employed to model turbulent combustion and NO formation process in practical engineering facilities to be adopted in the commercial software. In another attempt to numerically simulate NO formation, Guo et al. (2003) developed a modified version of the pre-PDF model, called the joint-PDF technique, for calculating the average rates of the chemical reactions. In their model, they benefited from the pre-PDF functions based on tridimensional Gaussian distributions of first and second orders to calculate the turbulent fluctuations of the reactant concentration and of the temperature, respectively. Their model was employed to predict the NO formation in a turbulent premixed combustion of methane and air coflow in sudden-expansion flow and diffusive turbulent jet. To close up the literature on numerical simulation of NO formation, a benchmark study had been carried out by Yang and Blasiak (2005) to numerically model the effect of the temperature on the combustion process of a preheated, non-premixed jet of propane and air coflow in a rectangular chamber. The numerical simulation was based on the average conservation equations of energy, momentum, and chemical species. Two combustion models were employed: the Eddy break-up (EBU) and the PDF with β function. The $k - \varepsilon - \mathrm{RNG}$ technique was used to solve the turbulence effects, while the NO formation was studied by considering two predominant NO formation mechanisms, i.e., the NO thermal and prompt mechanisms. Comparisons with the experimental data proved that the EBU model is more robust, especially with respect to the temperatures magnitudes and flame shape. Other important conclusion drawn from the study was that an increase in the fuel inlet temperature resulted in smaller flame, shorter mean residence time, smaller temperature peaks, and lower emission of NO.

In the current paper, a 2D numerical simulation of turbulent non-premixed coaxial methane combustion in the air enclosed by an axisymmetric cylindrical chamber is used to study the effects of species concentrations of reactants on the thermal and prompt NO formations, their individual contributions, and the chamber outlet temperature. A finite-volume staggered grid method is utilized to solve conservation equations of mass, energy, momentum, and species concentrations. Discrete transfer method (DTM) is used to solve radiation equation, and weighted-sum-of-gray-gases model (WSGGM), based on the newly obtained high-temperature molecular spectroscopic database (HITEMP)-2010 data, is utilized to account local variations of species absorption coefficients. To calculate NO concentration, a single- or joint-variable PDF in terms of a normalized temperature, mass fractions of species, or a combination of both is employed. The second-order upwind scheme is used for the space derivatives of the advection terms in all transport equations. The flow-field pressure linked equations are solved by coupled scheme to handle the velocity and pressure coupling. Also, a $k - \varepsilon$ model is employed to capture the turbulence in the chamber. The reaction is simulated using the eddy dissipation/finite rate model. This study had been carried from February 2014 at the institute of research for technology development (IR4TD).

Problem statement

The physical system considered here (Fig. 1) is the same case investigated in (Nieckele et al. 2001) and also studied as a case study in (Garréton and Simonin 1994). The axisymmetric cylindrical chamber schematized in Fig. 1 has a length and a diameter of 1.70 and 0.50 m, respectively. The fuel which in here mainly consists of CH_4 is injected into the chamber by a cylindrical duct aligned with the chamber centerline. In like manner, air is also injected coflow with the fuel. The fuel mass flow rates are 0.01453 and 0.1988 kg/s (with equivalence ratio of about 1.05) at temperatures of 313.15 and 323.15 K for fuel and air, respectively. For these mass flow rates, the corresponding velocities are 7.23 and 36.29 m/s for fuel and air, respectively. Thus, the Reynolds number at the entrance is about 18,000, emphasizing that the flow is turbulent. The fuel is injected into the chamber through a cylindrical duct with a spacing of 0.06 m, while air is brought into the chamber through a centered annular duct with a diameter of 0.02 m. The inlet air is composed of O_2, N_2, and H_2O (1 % in mass fraction), while the fuel is composed of CH_4 and of N_2.

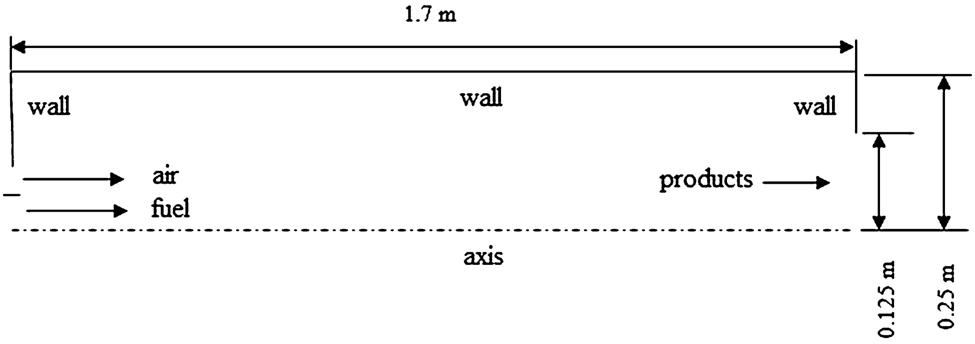

Fig. 1 Schematic of the axisymmetric cylindrical combustion chamber

Similar to the boundary conditions imposed by Garréton and Simonin (1994), in the current study, constant temperature (393.15 K) with no-slip conditions is considered for the combustion chamber walls. The walls are also assumed gray, diffusive emitters with an emissivity of 0.6. The inlet and the outlet reservoirs are assumed to act like black surfaces at the inlet and outlet temperatures. At the inlet, the velocity and concentration profiles are considered uniform. Pressure outlet boundary condition with zero gauge pressure is used for the outlet. The turbulence intensity is prescribed as 5 % for both the air and the fuel inlet flows. The burner power is about 600 kW. In the present study, the fan and the other external components are ignored in the computational domain, and their corresponding effects are considered through the inlet flow conditions.

Materials and methods

In this section, corresponding equations and the numerical solution procedure will be presented. In addition to the equation of state of ideal gas, the set of equations to be solved are mass, momentum, energy, and chemical species conservation equations. An Eulerian description for the fluid phase is adopted. The standard $k-\varepsilon$ turbulence model is employed to close the problem due to the averaged Navier–Stokes equation process.

The methane oxidation by atmospheric air was modeled by two global steps given by:

$$\begin{aligned}&2CH_4 + 0.22N_2 + 3(O_2 + 3.76N_2)\\&\quad \rightarrow 2CO + 4H_2O + 11.5N_2\end{aligned} \tag{1}$$

$$2CO + O_2 + 3.76N_2 \rightarrow 2CO_2 + 3.76N_2 \tag{2}$$

where the CO oxidation is modeled by the foregoing second reaction. The rate of formation or consumption, $R_{\alpha,c}$, of each αth species in each cth reaction (there are two reactions in the present study, so $c = 2$) is obtained by the Arrhenius-Magnussen's model.

Conservation equations in a general form can be written as (Centeno et al. 2014):

$$\frac{\partial}{\partial z}\left(\rho u\phi - \Gamma_\phi \frac{\partial \phi}{\partial z}\right) + \frac{1}{r}\frac{\partial}{\partial r}\left(r\rho v\phi - r\Gamma_\phi \frac{\partial \phi}{\partial r}\right) = S^\phi \tag{3}$$

in which ϕ, Γ_ϕ, and the source term S_ϕ can be determined using Table 1.

In Table 1, z and r are the axial and radial coordinates, u and v are the velocities in these respective directions, w is the angular velocity, ρ is the density of the gaseous mixture, μ is the gaseous mixture dynamic viscosity, and μ_t is the turbulent viscosity, defined as $\mu_t = C_\mu \rho k^2/\varepsilon$. The term $p^* = p - (2/3)k$ is the modified pressure, C_μ is an empirical constant of the turbulence model which is about 0.09, p is the combustion chamber operational pressure ($p = 101{,}325$ Pa), and k and ε are the turbulent kinetic energy and its dissipation. Also, $C_{1,\varepsilon}$ and $C_{2,\varepsilon}$ are empirical constants of the turbulence model ($C_{1,\varepsilon} = 1.44$ and $C_{2,\varepsilon} = 1.92$), and σ_k and σ_ε are the Prandtl numbers of the kinetic energy and dissipation, respectively ($\sigma_k = 1.0$ and $\sigma_\varepsilon = 1.3$). Pr_t and Sc_t are the turbulent Prandtl and Schmidt numbers, and R_α is the volumetric rate of formation or consumption of the αth chemical species (CH_4, O_2, CO_2, CO, H_2O). T is the temperature of the gaseous mixture. $C_{P,\alpha}$, h_α^0, and $T_{\mathrm{ref},\alpha}$ are the specific heat, the formation enthalpy, and the reference temperature of each αth chemical species. Finally, S_{rad} is the radiative heat source term.

In addition to the conservation laws presented in Table 1, an equation of state is needed to calculate the mixture density. Considering the mixture of fuel, oxidant and products as an ideal gas, the equation of state can be written as:

$$\rho = \frac{p}{\bar{R}T}\sum_\alpha \frac{y_\alpha}{MM_\alpha} \tag{4}$$

where $\bar{R}$ is the universal gas constant, MM_α is the mixture molecular mass, and y_α is the mean mass fraction of each αth chemical species.

Table 1 Definition of the terms in the generalized conservation equation, Eq. (3)

Equation	ϕ	Γ_ϕ	S_ϕ
Continuity	1	0	0
Axial momentum	u	$\mu + \mu_t$	$-\frac{\partial p^*}{\partial z} + \frac{\partial}{\partial z}\left(\mu_t \frac{\partial u}{\partial z}\right) + \frac{1}{r}\frac{\partial}{\partial r}\left(r\mu_t \frac{\partial v}{\partial z}\right)$
Radial momentum	v	$\mu + \mu_t$	$-\frac{\partial p^*}{\partial r} + \frac{\partial}{\partial z}\left(\mu_t \frac{\partial u}{\partial r}\right) + \frac{1}{r}\frac{\partial}{\partial r}\left(r\mu_t \frac{\partial v}{\partial r}\right) - \frac{(\mu+\mu_t)v}{r^2} + \frac{\rho w^2}{r^2}$
Turbulent kinetic energy	k	$\mu + \frac{\mu_t}{\sigma_k}$	$\left[\mu_t\left(2\left(\frac{\partial u}{\partial z}\right)^2 + \left(\frac{\partial u}{\partial r} + \frac{\partial v}{\partial z}\right)^2 + 2\left(\frac{\partial v}{\partial r}\right)^2 + 2\left(\frac{v}{r}\right)^2\right)\right] - \rho\varepsilon$
Turbulent kinetic energy dissipation	ε	$\mu + \frac{\mu_t}{\sigma_k}$	$C_{1,\varepsilon}\left[\mu_t\left(2\left(\frac{\partial u}{\partial z}\right)^2 + \left(\frac{\partial u}{\partial r} + \frac{\partial v}{\partial z}\right)^2 + 2\left(\frac{\partial v}{\partial r}\right)^2 + 2\left(\frac{v}{r}\right)^2\right)\right]\frac{\varepsilon}{k} - C_{2,\varepsilon}\frac{\varepsilon^2}{k}$
Energy	h	$\frac{\mu}{\mathrm{Pr}} + \frac{\mu_t}{\mathrm{Pr}_t}$	$S_{\mathrm{rad}} + \sum_\alpha \left[h_\alpha^0 + \int_{T_{\mathrm{ref},\alpha}}^{T} C_{p,\alpha}\mathrm{d}T\right] R_\alpha$
CH_4, O_2, CO_2, CO, and H_2O mass fraction	y_α	$\frac{\mu}{\mathrm{Sc}} + \frac{\mu_t}{\mathrm{Sc}_t}$	R_α

To consider thermal radiation exchanges inside the combustion chamber, the discrete transfer radiation model (DTRM) (Carvalho et al. 1991) is employed with the isotropic scattering media assumption. In the current study, the weighted factors polynomial coefficients and absorption coefficients are those considered in (Dorigon et al. 2013) for $p_{H_2O}/p_{CO_2} = 2.0$. Such WSGG correlations were fitted from HITEMP-2010, which is the most recent molecular spectroscopic database for high temperatures.

Discussed conservation equations are solved by spatial discretizing of each one in its dimensional version. The chosen function on the faces of the control volumes for diffusive–advective interpolation is the power law. Coupled method which is a pressure-based segregated algorithm is used to enable full pressure–velocity coupling. To give a summary of what the numerical simulation does, first from the problem specification, the thermo-physical properties of the involved species are obtained and from momentum equation, the velocity and pressure fields are then determined. In the next step, the enthalpy, chemical species concentration, and turbulence equations are discretized and solved. For the solution of the energy equation, the radiative heat source is first assumed null. After the temperature distribution is calculated, the radiative heat exchanges are computed incorporation coupled DTRM and WSGGM. This way, the radiative heat source is then obtained to be employed in the energy equation. To calculate NO concentration, a time-averaged NO formation rate is to be computed at each point in the domain using the averaged flow-field information. The method used for modeling the mean turbulent reaction rate here is based on PDF techniques (Liu et al. 2005). A typical run on an open-source software can take 5 h. It is worth noting that simulation is performed in a windows cluster on a system of 4 GB of RAM and CPU of 2.67 GHz.

Results and discussion

In this section, at first independency of the results with the employed mesh size is shown in Fig. 2. It is worth noting that the convergence criteria are determined by insuring that the maximum value of normalized residuals of any equation must be less than 10^{-6} except the NO formation which is 10^{-8}. As shown in grid independency test, Fig. 2, by employing 48,000 elements through the whole computational domain, one can make sure that the convergence criterion is favorably addressed. Second, validation task has been done by having a comparison between the experimental data from (Garréton and Simonin 1994) for similar case and conditions. As can be seen in Fig. 3, the O_2 mass fraction variation along the radial axis for the

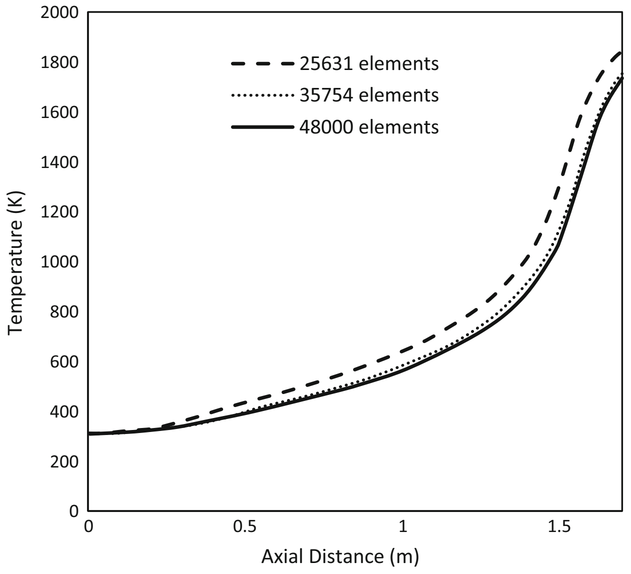

Fig. 2 Mesh size independency for the current numerical simulation: temperature along the axial distance

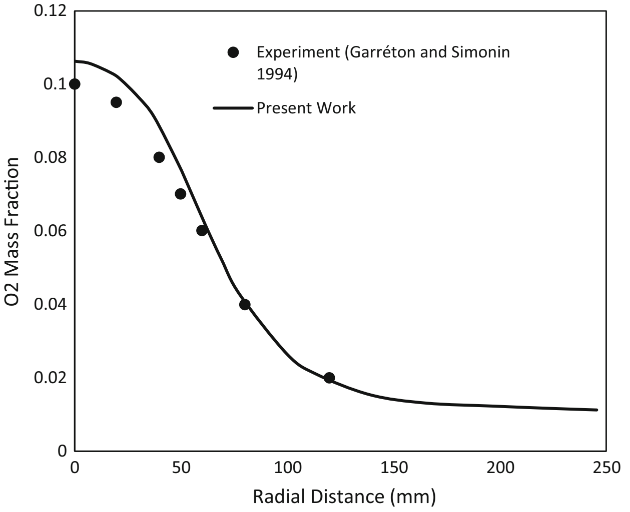

Fig. 3 Validation of the current numerical simulation: O_2 mass fraction along the radial axis

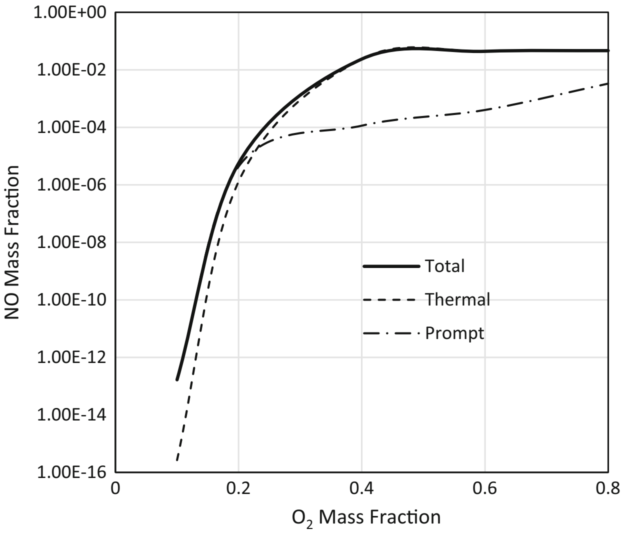

Fig. 4 NO mass fraction as a function of O_2 mass fraction

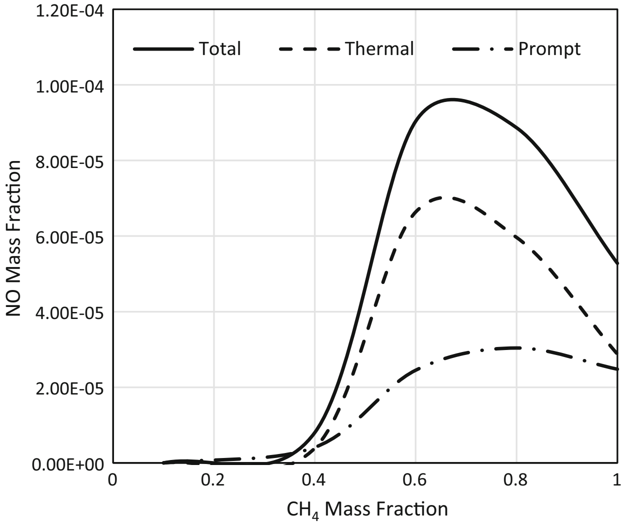

Fig. 5 NO mass fraction as a function of CH_4 mass fraction

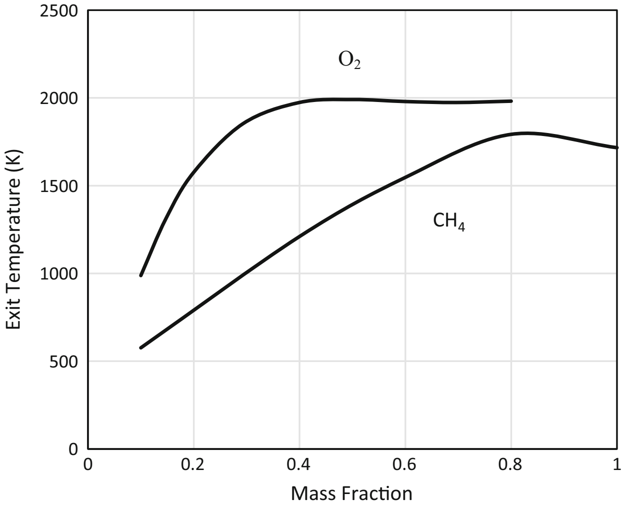

Fig. 6 Exit temperature as a function of CH_4 and O_2 mass fractions

current and the corresponding experimental works favorably matches. Third, total, thermal, and prompt NO formations with respect to the fuel and O_2 mass fraction variations are investigated in Figs. 4 and 5. It is important to note that the individual thermal and prompt NO mass fractions do not add up to the values predicted with the two models combined. The reason is that during combustion, reversible reactions take place and consequently NO produced in one reaction can be destroyed in another reaction. According to Fig. 4, up to 0.4 for O_2 mass fraction, NO formation substantially increases, and, however, after that, the NO mass fraction reaches a constant value. Besides, it shows that for the current case, the thermal NO formation mechanism is the dominant one determining the total NO formation. Figure 5 which presents NO mass fraction variations by CH_4 mass fraction, on the other hand, shows that both thermal and prompt mechanisms are contributing to the resultant total NO mass fraction. In this figure, parabolic shape of the curves, e.g., increasing and then decreasing trends, is because of the competitive nature of increasing fuel mass fraction and corresponding decrease in N_2 mass fraction for NO production. The figure also shows that by increasing fuel mass fraction, contribution of prompt and thermal mechanisms in NO production becomes approximately equal. Finally, the effects of mass fraction variations for CH_4 and O_2 on the exit temperature are investigated in Fig. 6. Calculated temperature is based on the mass-averaged outlet temperature computed as:

$$\bar{T} = \frac{\int T\rho\vec{v}\cdot \mathrm{d}\vec{A}}{\int \rho\vec{v}\cdot \mathrm{d}\vec{A}} \quad (5)$$

Apparent from Fig. 6, by increasing O_2 and CH_4 mass fractions, exit temperature measured at the outlet becomes nearly constant. The threshold values for these trends are 0.4 and 0.8 for O_2 and CH_4 mass fractions, respectively.

Conclusion

In the present work, a 2D numerical simulation of turbulent non-premixed coaxial methane combustion in air enclosed by an axisymmetric cylindrical chamber was carried out. A finite-volume staggered grid method was utilized to solve conservation equations of mass, energy, momentum, and species concentrations. Discrete transfer method (DTM) was used to solve radiation equation. Utilizing weighted-sum-of-gray-gases model (WSGGM), based on the newly obtained HITEMP-2010 data, local variations of species absorption coefficients were taken into account. The results proved that the employed numerical simulation can successfully predict species concentrations. Thereby, the numerical simulation was used to predict the effects of species concentrations of reactants on the thermal and prompt NO formations, their individual contributions, and the chamber outlet temperature. Investigating O_2 mass fraction effects on NO formation showed that after a specific value of O_2 mass fraction, NO formation would not change substantially. The same trend was seen for the exit temperature by increasing both O_2 and CH_4 mass fractions.

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