29th Congress of the International Council of the Aeronautical Sciences
St. Petersburg, Russia; September 7-12, 2014

MODELING STUDY OF POLLUTANT FORMATION IN HOMOGENEOUS METHANE AND SYNGAS FUELED COMBUSTOR

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Keywords: *modeling, homogeneous combustor, emission, methane, syngas*

Abstract

The comparison of emission characteristics of homogeneous methane- and syngas-fueled combustors is performed. Computations showed that replacement of methane by syngas allows burning much leaner fuel-air mixtures in homogeneous combustor with low values of NO and CO concentrations. However, in this case, combustion products contain notable amount of toxic gas N_2O and much greater amount of CO_2.

1 Introduction

Nowadays, one of the main goals in the combustion science is the decrease of the emissions of pollutants formed during combustion of fuel-air mixtures. The basic ecologically harmful gaseous substances formed at hydrocarbon fuel combustion are carbon (CO, CO_2) and nitrogen oxides (NO, NO_2). The decrease of CO emission is connected with the improvement of combustion process itself. It is more difficult to decrease the emission of NO. All proposed approaches come to decreasing the gas residence time in the high-temperature zone or to providing the reduced temperature in the combustion zone [1-8]. Today, one of the promising approaches to decrease the NO_x emission is the arrangement of stable burning of premixed homogeneous fuel-lean mixture. This is a reason why the creation of combustors with homogeneous burning mode attracts such great attention of the experts.

As the flame speed in syngas-air mixture (even at rather low percentage of H_2 in syngas H_2:CO=25:75) is considerably higher than that in methane-air mixture, the stable burning of syngas can be occurred in much leaner mixture. The NO concentration in combustion exhaust in this case can be potentially extremely low. This work is aimed to the modeling studies of emission characteristics of homogeneous combustors using methane and syngas as a fuel.

2 Kinetic model

The reaction mechanism of methane oxidation used for modeling in the present work was taken from [9]. It includes also submechanism of syngas oxidation. This mechanism was validated on a vast set of experimental data on the ignition delay time, velocity of laminar flame propagation, and evolution of gas temperature and species concentrations in the flame front of methane–air and syngas–air mixtures [9, 10]. This reaction mechanism was supplemented with the block of reactions involving following N-containing species: N, N_2, NO_x (x=1...3), N_2O, HNO_x, HCN, CN, CNO, NH_x and N_2H_y (y=1...4) and including principal mechanisms of NO formation: thermal (or extended Zel'dovich) mechanism, Fenimore's (or prompt) mechanism, N_2O-mechanism, and NNH-mechanism [11].

The kinetic model was validated against the profiles of NO concentration in a plane laminar methane–air flame [12] and demonstrated close fit to experimental data for stoichiometric and fuel-lean mixtures [13].

The measurements of NO and CO concentrations were also carried out for Bunsen burner operating on methane at atmospheric

conditions (the pressure and temperature were P_0=l atm and T_0=300 K) [14]. The burner was a vertical cylindrical tube with the length 82 cm and inner diameter 17 mm. Such a long tube provided a completely developed laminar flow at the exit. The flow rates of methane and air were 0.97 and 6.69 liter/min, respectively, i.e. a fuel-rich methane–air mixture was used (fuel/air equivalence ratio ϕ was equal to 1.38). The concentrations of NO, CO, and OH, and the temperature were measured by the laser induced fluorescence (LIF) and coherent anti-Raman scattering (CARS) techniques at cross sections of the flame at several distances from the burner exit. In order to validate our kinetic model the numerical simulation of this experiment was performed with the use of the FLUENT code of the ANSYS-CFD package [15].

Figures 1 and 2 show the experimental and calculated distributions of temperature and mole fractions of NO and CO over the transversal coordinate in the cross section located at the distance y=21 mm from the burner exit. Calculations were carried out with the use of the kinetic model of this work and the reaction mechanism GRI-Mech 2.11 [16]. Both reaction mechanisms adequately describe the experimental profiles of temperature and concentrations of CO. A small difference of calculated profiles from measured values of temperature and CO mole fractions can be connected with the some uncertainties in the choice of boundary conditions during the simulations. However, the NO mole fraction profiles obtained with use of these two reaction mechanisms are essentially different. The kinetic model of this work describes the experimental data rather correctly, whereas the GRI-Mech 2.11 reaction mechanism predicts twofold overestimation of the NO mole fraction compared to experiment.

Thus, the kinetic model of this work ensures an adequate description of the measured concentrations of NO and CO in laminar flames. Let us analyze now the formation of environmentally harmful species in homogeneous combustors operating on fuel-lean methane-air and syngas-air mixtures.

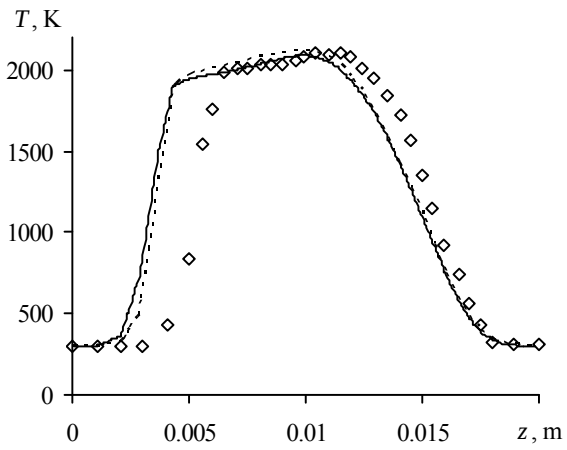

Fig. 1. Distribution of temperature in the flame cross section located at a distance y=21 mm from the Bunsen burner exit: the symbols are the experimental data [14], the dashed and solid curves are the predictions of reaction mechanism [16] and kinetic model of this work.

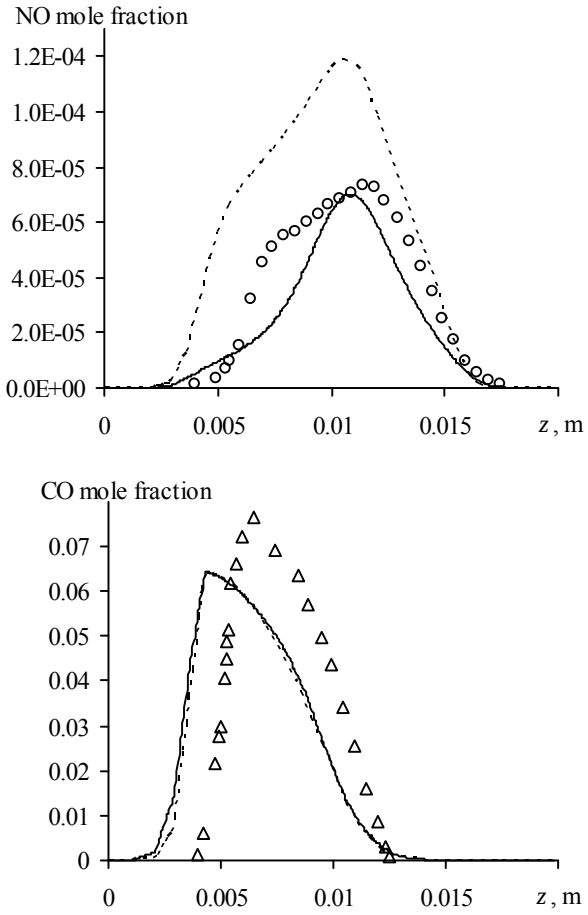

Fig. 2. Distributions of NO and CO mole fractions in the flame cross section located at a distance y=21 mm from the Bunsen burner exit: symbols are the experimental data [14], the dashed and solid curves are the predictions of reaction mechanism [16] and kinetic model of this work.

3 Metodology

Modeling of fuel burning in the combustor was carried out with the use of FLUENT program from ANSYS-CFD package [15]. During modeling the processes in a combustor, it is needed to choose a correct model of combustion. Within the framework of FLUENT code, there are two combustion models that enable one to use the detailed reaction mechanisms: quasi-laminar model and model of eddy-dissipation. For the choose of combustion model that describes the processes in the homogeneous combustor more accurately, the comparison of the predicted fields of OH concentration with the use of quasi-laminar and eddy-dissipation-concept models with the measurements of OH concentration field [17] has been performed.

Fig. 3 shows the geometry of the combustor taken from [17]. The cylindrical wall of the combustor was cooled by air. In accordance with brief description of experimental setup, the temperature in our calculations was taken 807 K in the entrance cross section, 770 K on the left (entrance) and 300 K on the right (exit) boundaries of the cylindrical wall. The temperature was assumed to change linearly from the left to the right end of the combustor. The pressure, temperature and velocity of the homogeneous mixture at the combustor input were P_0=5 bar, T_0=673 K, u_0=45 m/s. The homogeneous syngas-air mixture H_2/CO=1/1 with ϕ=0.54 was used. Taking into account that the flow in the combustor is axisymmetric, an approach based on the averaged Reynolds equations and the famous two-parameter k–ε turbulence model was used. The parameters of the turbulence (turbulence energy and its dissipation rate) at the combustor entrance were determined from the condition that the intensity of velocity fluctuations was 10% and the integral scale of turbulence was equal to 1.2 mm.

The OH concentration fields measured in the experiment [17] and calculated by these two combustion models are shown in Fig. 4. One can see that the length of the jet core calculated by the quasi-laminar model is close to experimental length. At the same time, the

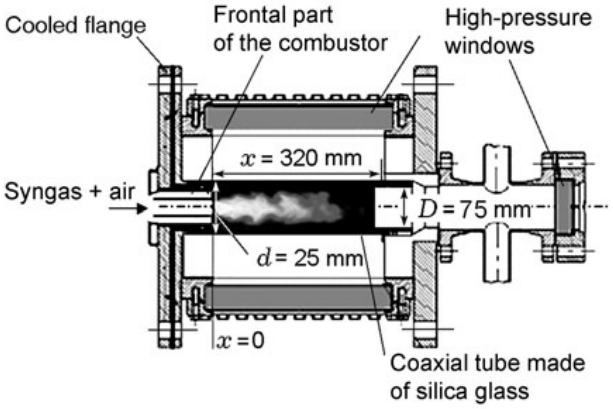

Fig. 3. Geometry of a homogeneous combustor.

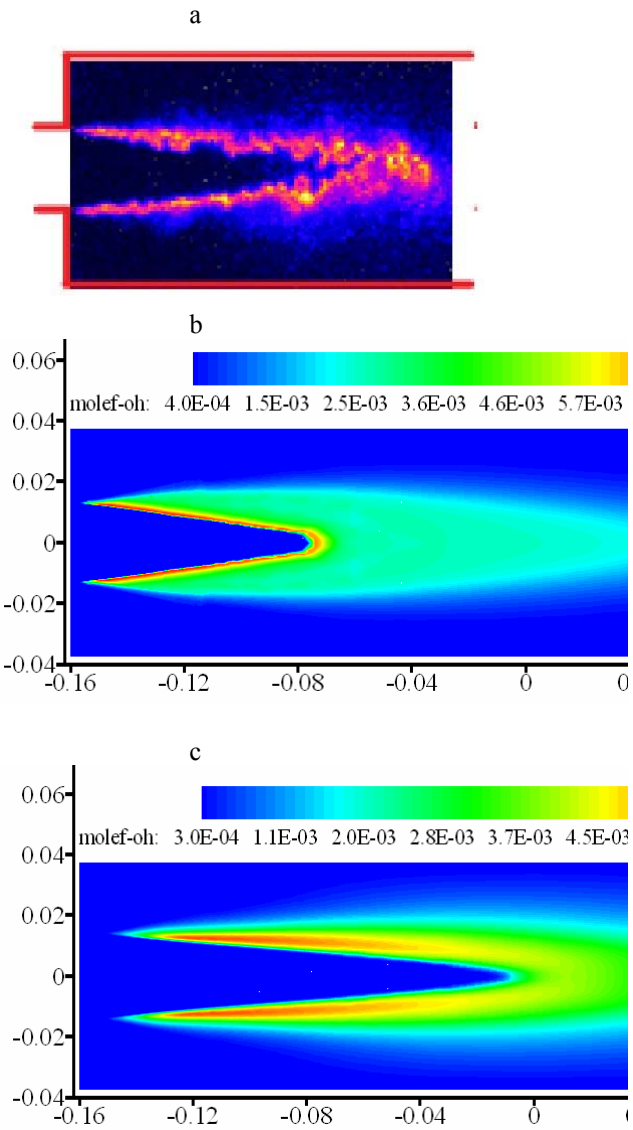

Fig. 4. The field of OH radical measured in [17] (a) and calculated by the quasi-laminar combustion model (b) and by the model of dissipation of vortices (c) in a homogeneous combustor operation on syngas (ϕ=0.54).

model of eddy-dissipation predicts longer jet core. It means, that quasi-laminar combustion model allows one to provide good agreement with the experiment.

Therefore, all further calculations of emission characteristics were conducted with the use of quasi-laminar combustion model. For the calculation of NO concentration the post-processor procedure of FLUENT program was used.

4 Modeling and Results

The measurements of CO and NO concentrations were performed in [17] for the same combustor (see fig. 3). Combustion of syngas (H_2/CO=1/2) in air with ϕ=0.33 was studied in these experiments. The measured values of the CO and NO concentrations at the combustor exit were 0 and 1.2 ppm, respectively [17]. Calculations gave the values: 0.0001 ppm for CO and 3.8 ppm for NO (at 15% of O_2), that are in the reasonable consistence with experimental data. From these data one can see that burning the ultralean homogeneous syngas–air mixture ensures extremely low concentrations of CO and NO in homogeneous combustor. However, the calculations predict that the combustion products also contain 2.5 ppm N_2O that is comparable with NO concentration. It should remind that nitric oxide is a toxic gas and more dangerous for people and animals than NO.

Let us compare the emission characteristics of the homogeneous combustor when using different fuels: methane and syngas. A particular analysis was performed for the homogeneous combustor [18]. This combustion chamber worked without cooling at pressure P_0=1 atm and initial mixture temperature T_0=300 K. The chamber represents two cylindrical tubes with diameters of 65 and 40 mm attached to each other. The length of the tube with larger diameter was equal to 30 cm, and diameter of entrance orifice was equal to 20 mm. The residence time in the chamber was approximately equal to τ=32 ms. According to [18], the methane-fueled combustor provides stable burning of the fuel-lean mixture at ϕ=0.65-0.95. Therefore, the calculation of combustion of homogeneous methane-air mixture was carried out at ϕ=0.73. Calculations of syngas-air mixture burning (H_2:CO=1:1) were performed at several values of ϕ.

The results of computations presented in the Table show that the replacement of methane by syngas at ϕ=0.73 leads to more than tenfold growth of NO and CO emissions. However it is possible to burn the syngas at twice smaller value of ϕ (ϕ=0.365) at which NO and CO concentrations in the combustor exit are small (not more than 3 ppm at 15% O_2). Note, that methane-air mixture cannot burn stably at ϕ=0.365. Thus, the replacement of methane by syngas allows to burn leaner mixture in homogeneous combustor with low values of NO and CO emissions. However, in this case, the combustion exhaust comprises notable amount of N_2O that is strongly toxic gas. In addition, the concentration of CO_2 (it is greenhouse gas) increases by a factor of 2 in the combustion products.

Table. Temperature and species concentrations [ppm 15%O_2] polluting the atmosphere on the exit of homogeneous combustion chamber when methane or synthesis-gas are used as fuel.

	CH_4/air	H_2/CO/air		
	ϕ=0.73	ϕ=0.73	ϕ=0.63	ϕ=0.365
T_{ex}, K	1962	2121	1963	1517
NO	16.1	370	66.4	0.57
N_2O	0.07	0.27	0.26	0.55
CO	137	1787	459	2.7
CO_2	33406	58300	59108	64605

Compare the emission characteristics of the combustor operating on methane and syngas when identical temperatures of the combustion products T_{ex} are realized at combustor exit. Calculations show (see Table) that the identical value of T_{ex} is reached by burning a methane–air mixture with ϕ=0.73 and a syngas–air mixture with ϕ=0.63. In the case of syngas burning, the concentrations of all considered environmentally harmful species at the combustor exit are much higher (by a factor of 2 and more) than in the case of burning the methane–air mixture. Thus, in the case of identical thermodynamic efficiency, syngas

does not ensure any advantages over methane in terms of a more environmentally clean exhaust.

5 Conclusions

The developed detailed kinetic model provides an adequate description of experimental profiles of NO concentration along the front of the laminar atmospheric methane–air flame and the transversal profiles of temperature, CO and NO concentrations measured in the Bunsen burner. This model correctly predicts the CO and NO concentrations at the exit of homogeneous combustor. This is possible only by using the quasi-laminar combustion model.

The use of syngas instead of methane in the homogeneous combustor operating on a fuel-lean mixture leads (at an identical ϕ value) to a significant increase (up to 20 times) in the emissions of CO and NO, as well as the toxic gas N_2O (by a factor of 4) and to a noticeable increase (by a factor of 1.7) in the CO_2 concentration in the exhaust gases. However, the use of syngas allows one to arrange stable combustion of much leaner mixture than that in the case of the usage of methane and to obtain lower emissions of CO and NO. At the same time, burning of such fuel-lean mixtures yields higher concentrations of N_2O in combustion products. Therefore, the statement of obtaining an environmentally clean exhaust by using syngas as a fuel is not justified.

6 Acknowledgements

The work was supported by the Russian Foundation for Basic Research (Grant Nos. 14-08-00794 and 13-08-90441) and by the Grant of the President of the Russian Federation for Supporting Young Scientists and Leading Scientific Schools (Grant No. NSh-960.2014.8).

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