

Numerical and Experimental Investigations
on Reduction of NO and CO Emissions
in City Gas Combustion

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Chapter 1: Introduction

1.1 Background and motivation for this study

Carbon dioxide (CO₂) emissions, which are thought to cause global warming, are increasing at a rapid rate. The Greenhouse Gas Inventory Office of Japan (GIO) annually reports the National GHG Inventory Report of JAPAN [1]. The breakdown of CO₂ emissions in FY2018 shows that the emissions from fuel combustion were the highest at 94.9%. Looking at the breakdown of the emissions due to fuel combustion, the energy conversion sector such as power generation plants accounted for 41.6%, the manufacturing sector accounted for 23.1%, and the transportation sector accounted for 17.9%. The CO₂ emissions in the energy conversion sector decreased by 7.1% compared to that of the previous year, but increased by 28.2% from FY1990. The reason of the increase is that the solid fuel consumption in power generation plants increased. On the other hand, the CO₂ emissions in the manufacturing sector decreased by 2.6% compared to that of the previous year, and decreased by 24.8% from FY1990. One of reasons of the decrease is that fuel conversion from heavy oil to city gas, which is mainly produced from natural gas, reduced liquid fuel consumption. The carbon emission factors for each fuel type in 2018 were 24.6 t-C/TJ for steel making coal, 19.3 t-C/TJ for crude oil for power generation use, and 13.9 t-C/TJ for liquefied natural gas (LNG) [2]. Thus, CO₂ emissions from natural gas are 43% less than those from coal and 28% less than those from oil. As mentioned above, to reduce the CO₂ emissions from fuel combustion, fuel conversion such as from heavy oil to city gas would be an effective approach.

To further reduce CO₂ emissions, the following measures are important. First, in the energy conversion sector, it is necessary to utilize more renewable energy sources such as solar power and wind power, and to develop electricity storage technologies to address their intermittency variability problems. However, from the viewpoint of supply stability and environmental compatibility, these technologies are not sufficient and it is necessary to continuously study the best mix with thermal power generation. Among the thermal power generation technologies, the demand for gas turbine power generation, which emits less CO₂, will increase for the future. In Japan, where natural resources are scarce, most of the natural gas is imported as LNG. Therefore, the equipment that can

stably vaporize a large amount of LNG is required, and increasing the size of the Submerged Combustion Vaporizer (SCV), which is one of the types of LNG vaporization equipment, is important [3]. The SCV system is a facility that burns a part of the vaporized fuel and stably vaporizes LNG by combustion heat [4]. Large-scale SCVs are required not only in Japan but also in Asian countries such as South Korea and China, and this development can promote the spread of clean natural gas worldwide. In the medium term, it is necessary to introduce fuel that does not emit CO₂ into gas turbine power generation. Hydrogen and ammonia (NH₃) are examples of such fuel candidates. Mitsubishi Heavy Industries, Ltd. is developing a hydrogen gas turbine, and plans to introduce a 100% hydrogen burning facility to the Netherlands by 2025 [5]. On the other hand, in the manufacturing sector, the conversion from heavy oil fuel to city gas needs to be continued and accelerated, and in the future, as in the energy conversion sector, it is effective to promote the use of ammonia, which is a fuel that does not emit CO₂.

Summarizing the above, several challenges regarding combustion technologies need to be addressed in chronological order. Within the most recent years up to 2020, the development of an large-scale SCV burner with the large vaporization capacity of 200 t/h is required [2, 4, 6, 7]. In the medium term up to 2030, adding ammonia as a fuel for gas turbine power generation to reduce CO₂ emissions will be tested [8]. Furthermore, in the long-term up to 2050, we will also introduce ammonia as fuel in the manufacturing sector, aiming to achieve both high thermal efficiency and low emission of harmful substances [9].

There are many difficulties in developing combustors which meet the above requirements with various combustion modes depending on the application. The larger the burner, the more limited the information obtained from combustion experiments, and it is uneconomical to optimize the design through numerous experiments. Therefore, performing numerical simulations of combustion fields has become important for optimizing the burner structure. With the development of simulation technologies in recent years, it has become possible to perform numerical simulations of combustion fields targeting various actual combustors that could not be performed in the past [10]. Further, new combustor designs should reduce harmful combustion emissions as much as possible. The main emissions are nitric oxide (NO) and carbon monoxide (CO). NO is one of the nitrogen oxide (NO_x), which is a causative agent of photochemical smog and

acid rain, and has an adverse effect on the human health. On the other hand, CO has serious effects on the human health. Therefore, it is important to predict both NO and CO emissions in the combustors by the numerical simulations of combustion fields.

Therefore, the goal of this study is to develop the numerical simulation methods for the combustion fields which can simulate for realistic combustors and predict their NO and CO emissions with high accuracy. In this thesis, three combustors are targeted: the first is large-scale SCV burners which have water spray system for suppressing NO production, the second is a laboratory scale gas turbine combustor which uses the mixture of ammonia and methane as fuel, and the third is Moderate or Intense Low-oxygen Dilution (MILD) combustion [11], which is mainly used technique in the manufacturing sector, of ammonia and city gas. As a numerical simulation method, Large Eddy Simulation (LES) is mainly employed. The computational load of LES is higher than that of the Reynolds-Averaged Navier-Stokes (RANS) simulation, which adopts time-averaged models and widely-used in industries, but vortices larger than the spatially filtered scale are directly calculated, resulting in a more accurate solution. As turbulent combustion models, the non-adiabatic flamelet methods constructed based on Flamelet Progress Variable (FPV) [12] or Flamelet Generated Manifold (FGM) [13] are used. The flamelet libraries are generated by using the calculation results of the one-dimensional flames considering a detailed chemical reaction mechanism. By considering the enthalpy loss from the adiabatic state, the change in the chemical reactions due to the heat loss can be considered. This is because actual combustors involve the heat losses due to radiation and convection to the cold wall of the combustor. In particular, the reactions of NO and CO are highly temperature-dependent, so it is predicted that handling of this non-adiabatic state will be extremely important. In principle, in the flamelet library, the chemical species concentrations are stored as equilibrium states. Since the production and consumption rates of NO and CO are much slower than the reaction rates of major species, the approach of Ihme and Pitsch [14] is adopted, and the production and consumption rates of NO and CO stored in the flamelet library are used for their transportation, and the emissions are predicted through solving the equation. However, only in Chapter 4, since the MILD combustion that is the subject of research cannot be expressed by the flamelet methods, the Eddy Dissipation Concept (EDC) model is used as a turbulent combustion model [15]. Also, in order to compare the NO and CO emissions predicted by the

numerical simulations of the MILD combustion fields, the combustion experiments will be conducted for the water spray turbulent flames of the SCV burners and MILD combustion. The swirl flow flame of ammonia and methane in Chapter 3 will be compared with the experimental results in previous studies.

1.2 Thesis outline

Figure 1.1 shows the structure of this thesis. This chapter is Introduction, and generally gives the description of research background and motivation of this study. Three combustion fields are studied in Chapters 2-4, respectively: the first is a turbulent flame of city gas/air with water spray in SCV burners, the second is a turbulent premixed flame of ammonia/methane/air in a laboratory scale swirl combustor, and the third is MILD combustion of ammonia/city gas/air in a high temperature air furnace. When classified by fuel type, the fuel in Chapter 2 is city gas, and the fuel in Chapters 3-4 is the mixture of ammonia/hydrocarbon, that is, ammonia/methane or ammonia/city gas. When classified according to the turbulence combustion model used, that used in Chapters 2-3

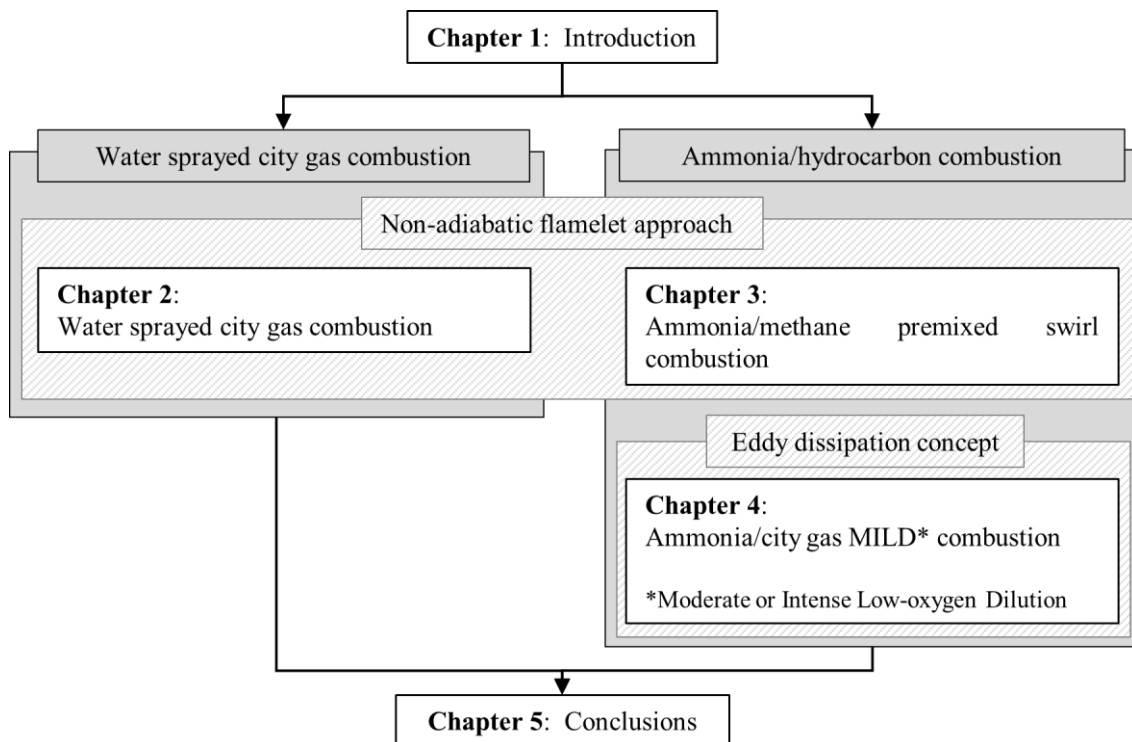


Figure 1.1 The structure of this thesis.

is the various non-adiabatic flamelet approaches, and that used in Chapter 4 is the EDC model.

In Chapter 2, the turbulent flames with water spray in the SCV burners are focused on. There are three objectives in this chapter. The first objective is to develop the large-scale SCV burner through numerical simulations and experiments. As a turbulence combustion model for the LES, Non-Adiabatic Flamelet Progress Variable (NA-FPV) is developed based on the conventional Flamelet Progress Variable (FPV) [4]. This burner has water spray system to suppress NO production. The consideration of heat losses by the NA-FPV is effective for expressing the heat loss by water evaporation. Additionally, in order to consider the effect of the dilution by water vapor, water vapor concentration, W , is added as a new control parameter. The second is to compare numerical simulation results using the non-adiabatic flamelet methods of the premixed flame type (Flamelet Generated Manifold, FGM) and diffusion flame type to find a more suitable method for predicting NO and CO emissions [6]. This study is important to understand which premixed flame or diffusion flame characteristics dominate a real combustor. There are three governing parameters in the conventional FPV or FGM, whereas there are five governing parameters in this study. The increase of the governing parameter causes an explosion of the data capacity of the flamelet library and limits the high resolution. This is a factor that reduces the calculation accuracy. Then, the third is in order to improve the accuracy of the high-dimensional flamelet approach and expand the scope of application, to perform neural network modeling by the deep learning technique and verify the calculation accuracy.

In Chapter 3, the turbulent premixed flames of ammonia/methane/air in a laboratory scale swirl combustor are focused on. The objective is to predict NO and CO emissions through the LES. The mixture of ammonia/methane/air is supplied, so Non-Adiabatic Flamelet Generated Manifold (NA-FGM) based on FGM is used as the turbulence combustion model [8]. Also, the reaction characteristics of NO and CO in ammonia/methane co-combustion are also investigated through zero-dimensional calculation using the detailed chemical reaction mechanism.

In Chapter 4, the MILD combustion of ammonia/city gas/air in a high temperature air furnace is focused on. Flamelet approach cannot be applied to MILD combustion with unclear flame surface. Therefore, numerical simulations of the MILD combustion fields

here are performed using the RANS simulation employing the Eddy Dissipation Concept (EDC) model with the detailed chemical reaction mechanism [9]. After verifying the accuracy of numerical simulations of MILD combustion fields of city gas/air through the comparison with the experimental results, numerical simulations of ammonia/city gas/air MILD combustion field will be carried out. The objective is to investigate important burner design parameters for reducing NO emissions.

In Chapter 5, the conclusions are summarized through the whole thesis.

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Chapter 2: Water sprayed city gas combustion

2.1 Introduction

The demand for natural gas, which has a low environmental impact among fossil fuels, is increasing worldwide. Countries with limited natural gas resources, including Japan, import natural gas through pipelines or liquefied natural gas (LNG) tankers. In 1969, Tokyo Gas, together with Tokyo Electric Power Company (TEPCO), procured LNG through a tanker for the first time in Japan [1]. Japan is currently the world's largest LNG importer, having imported about 30% of the world's total imports as of 2016 [2]. In 2025, the global demand for LNG trade is expected to double due to the rising demand in developing countries [3].

After being imported by an LNG tanker, the LNG is vaporized at a terminal, using two vaporization methods: an open rack-type vaporizer (ORV), which vaporizes by providing considerable seawater to the heat exchanger through which LNG is passing, and a submerged combustion vaporizer (SCV), which burns fuel gas in a burner immersed in water and vaporizes LNG through the heat of combustion. SCV is an indispensable facility for LNG terminals that cannot install ORVs, which require considerable water, because LNG can be stably vaporized by burning fuel gas. Figure 2.1 shows the schematic of an SCV system. It comprises a fuel supply device, a blower for supplying combustion air, a submerged burner, a water pool for transferring heat, and heat transfer tubes. SCV has the following advantages over ORV. (1) It can be used at terminals where an ORV cannot be installed, such as inland areas where enough seawater cannot be obtained and cold areas where seawater temperature is low. (2) Its evaporation capacity per installation area is larger than that of an ORV, because a high-temperature burned gas is used as the heat medium. (3) Even if the fuel gas supply suddenly stops, LNG can be vaporized for a certain period using hot water stored in the water pool. (4) Because intake and drainage equipment, such as an ORV, are not required, the construction cost is low. However, as its disadvantages, (1) the operating costs are high because about 1.5% of the LNG vaporization capacity is consumed as fuel and (2) compliance with environmental regulations is required to release combustion exhaust gases, especially nitrogen oxide

(NO_x).

To decrease the fuel cost, some previous studies have provided useful information about the characteristics of heat transfer in SCVs. Based on computational fluid dynamics (CFD) and theoretical analysis, a systematic and effective method to describe the heat transfer for designing practical systems has been proposed [4, 5]. In addition, the characteristics of heat transfer have been numerically investigated, and it has been found that the water temperature decreases with LNG inlet temperature and increases with LNG mass flow rate and tube fouling resistance [6].

Besides heat transfer characteristics, there is another important challenge for SCV development, namely the reduction in combustion emissions such as NO_x and carbon monoxide (CO) in the burner. To the best of our knowledge, only a few studies have reported combustion emissions in SCV burners. Tokyo Gas Engineering Solutions (TGES), in collaboration with Tokyo Gas and Sumitomo Precision Industries, has worked on SCV burner development for many years. Figure 2.2 shows the change in the cumulative number of SCV burners shipped by TGES. In recent years, many SCV burners have been manufactured and exported for not only Japanese terminals but also overseas terminals. In addition, due to the growing demand for natural gas, the vaporization capacity, which was about 50 t/h 40 years ago, has increased to 200 t/h. As SCV burner

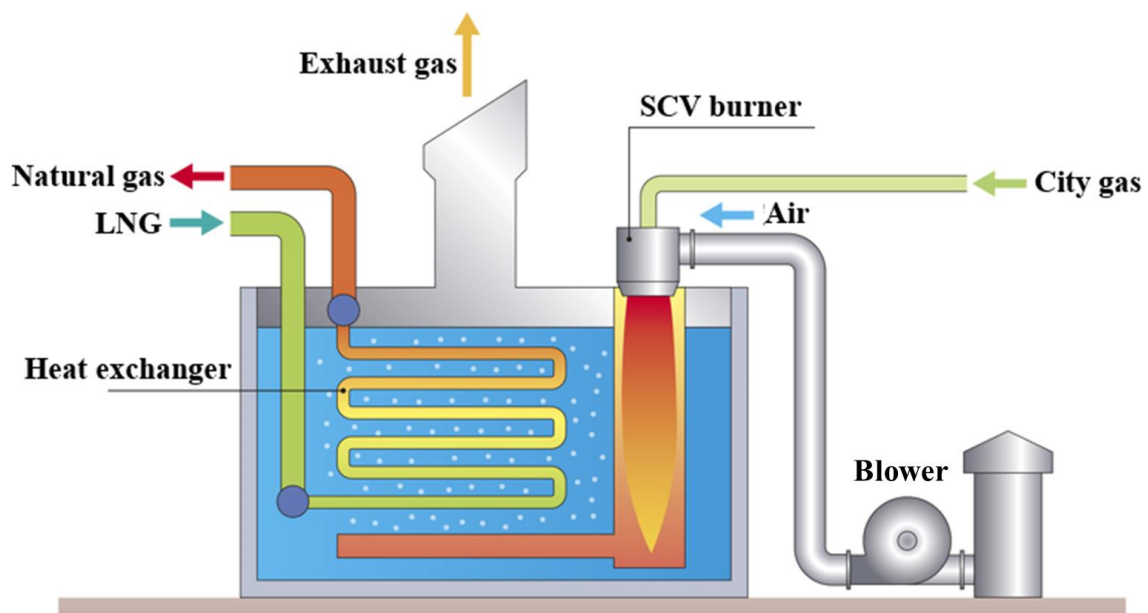


Figure 2.1 The structure of SCV burner.

2.1 Introduction

has a special usage environment as combustor, it cannot easily control NO_x and CO emissions. First, as the combustion chamber becomes large, the calorific value per volume increases and NO_x is easily generated. Therefore, our final goal is to reduce NO_x production by installing a water spray system [7]. The suppression of NO_x formation through water spray is well known [8]. In general, the evaporation latent heat loss leads to a decrease in system efficiency, and thus, there are few examples where water spray systems are introduced into practical combustion systems. In an SCV, the exhaust gas temperature decreases to ambient temperature by exchanging heat with the water of a heat medium and thus, the thermal efficiency hardly deteriorates even though water is sprayed. For this reason, the introduction of a water spray system is a reasonable method for directly suppressing NO_x production for SCV burners. On the other hand, considerable CO is likely to be emitted due to the following three major factors. (1) The flame is rapidly cooled by the latent heat loss of water evaporation. (2) Because the combustion chamber is immersed in water of less than 100°C, the flame is cooled by radiation and convection to the cold chamber walls. (3) To maintain the amount of burned gas to be bubbled into the water pool, the amount of air becomes too excessive depending on the combustion load and burner structure.

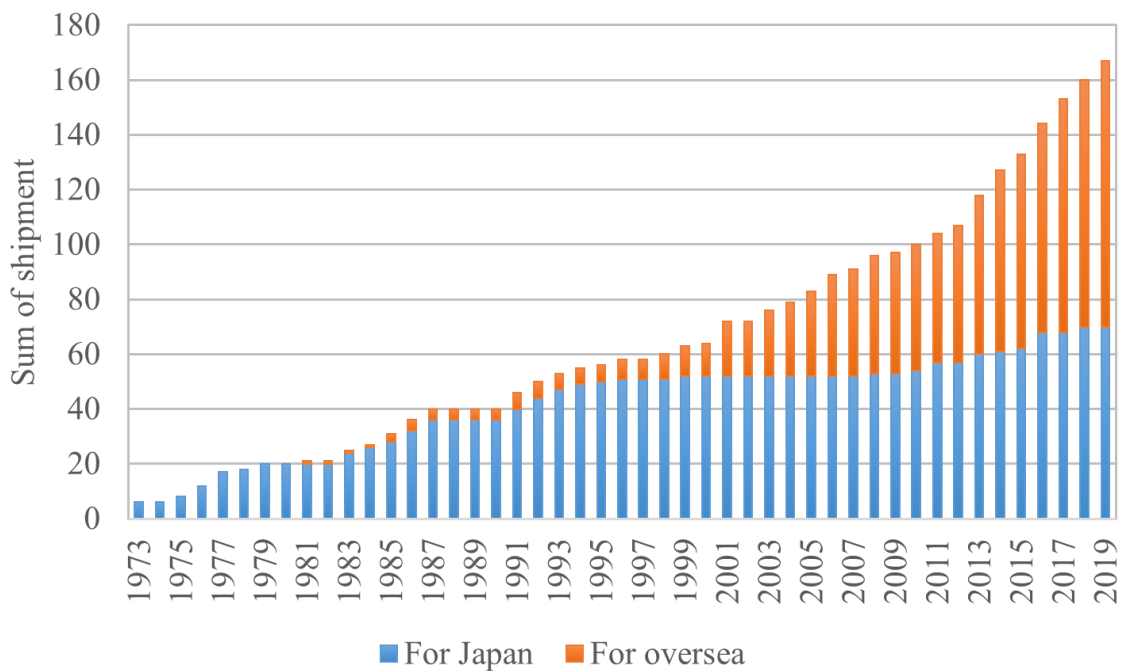


Figure 2.2 Comericial shipment of SCV burner.

This chapter has three purposes. The first purpose is to develop a large-scale SCV burner equipped with a water spray system by numerical simulations of cold flows, and to investigate the characteristics of the NO_x and CO emissions in detail. In addition, Large Eddy Simulations (LESs) of turbulent combustion fields generated by the large-scale SCV burner are performed and the validity is assessed by comparing with the experiments. As a combustion model, the Five-Dimensional Non-Adiabatic Flamelet Progress Variable (5D-NA-FPV) approach is newly introduced [7]. The 5D-NA-FPV is based on Flamelet Progress-Variable approach (FPV [9]) whose library is generated from counter flow diffusion flame calculations. This approach can take into account the effects of various heat losses, including latent heats by the evaporation of sprayed water, conduction, and radiation of hot burned gases, on the reactions of nitric oxide (NO) and CO.

The second purpose is to compare LESs employing two kinds of non-adiabatic flamelet approaches, namely the non-adiabatic premixed and non-premixed flamelet approaches which are referred to as 5D-NA-FGM [10] and 5D-NA-FPV [7, 10, 11], respectively. The 5D-NA-FGM is based on Flamelet Generated Manifold approach (FGM [12]) whose library is generated from one-dimensional free-propagating flame calculations. For both approaches, in order to consider the heat loss due to water evaporation and the addition of water vapor to mixed gases, the enthalpy drop from adiabatic state and the water vapor mass fraction are taken into account in the flamelet libraries. This study is conducted for a medium-scale SCV burner.

The third purpose is to develop a neural network model based on the 5D-NA-FGM in order to reduce the library sizes. The 5D-NA-FGM and 5D-NA-FPV have too large library's size due to having many control variables with high resolutions. In general, computers have several GBs of memory; however, if the number of control variables is more than four, the database size increases to tens of GBs, thereby making it impossible to employ the high-dimensional flamelet database on computers. Recently, the compression of these databases through the use of deep learning has been investigated in a few studies [13, 14]. In this study, a five-dimensional flamelet database is created to consider the effects of not only heat losses but also the addition of water vapor generated by the evaporation of sprayed water. The neural network is learned by using the database as a teacher dataset. LESs for a small-scale SCV burner are conducted using this model, and the behavior is investigated in detail. For comparison, the burner equipped with a

water spray nozzle is manufactured, and combustion experiments with and without water spray are also conducted.

2.2 Structures of SCV burners

In this chapter, SCV burners with three different sizes are studied. The following sections describe the structures of each burner.

2.2.1 Large-scale burner

Figure 2.3 shows (a) the conventional structure of a large-scale SCV burner, and the newly proposed (b) swirl flow structure of that. The fuel supply pipe system and water spray system are common for both structures. The fuel is supplied from the upper part, branched into eight pipes inside the burner, and then injected into the combustion chamber. The water spray nozzle is mounted in the upper center inside the burner and sprays water downwards. The air flow concepts for each structure differ, as described below. (a) The conventional structure is the simplest structure studied in early design. The air supplied

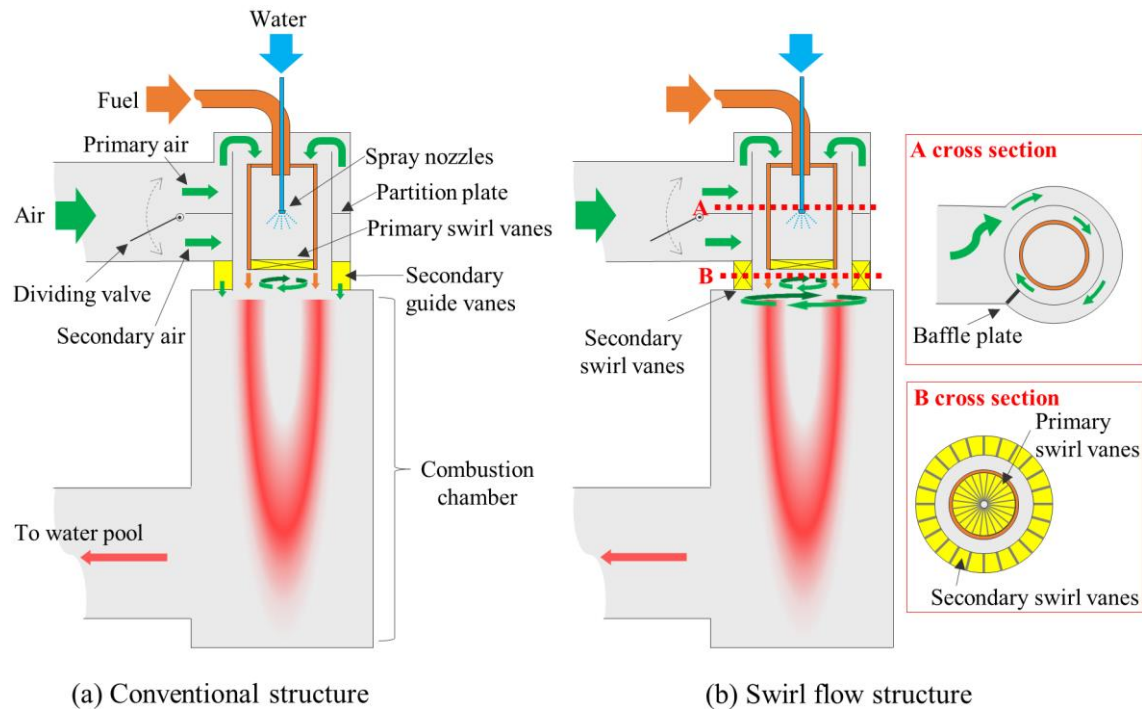


Figure 2.3 Schematics of large-scale SCV burners for (a) conventional structure and (b) swirl flow structure.

by the blower is distributed to the primary and secondary air by using a dividing valve. The primary air flows inside the burner and forms a swirl flow by primary swirl vanes, and then, mixes with the fuel and reacts. The secondary air is rectified by secondary guide vanes, whose inclination angle is zero, and then, supplied to the combustion chamber. To vaporize stably LNG, the flow rate of the burned gas is maintained constant as the combustion load decreases. Therefore, in that case, the primary air rate decreases, while the second air rate increases. That is, the total excess air ratio, λ , changes depending on the combustion load, as follows: $\lambda = 1.3$ at 100% load, $\lambda = 2.6$ at 50% load, and $\lambda = 12$ at 10% load. In addition, the burned gas produced in the combustion chamber flows into a sparge pipe that has many small holes for the generation of bubbles. (b) The swirl flow structure has been introduced with the aim of making the flow field uniform while maintaining the outer dimensions of the conventional structure. By arranging a baffle plate on the upper part of the burner, the primary and secondary air are swirled in advance and a uniform swirl flow is formed via the primary and secondary swirl vanes.

The structure of the burner with the swirl flow structure has designed through numerical simulations of cold flows. Figure 2.4 shows the computational domain for the conventional structure. The rated LNG vaporization capacity is assumed to be 200 t/h. Here, the structure is exquisitely reproduced to focus on the air flow, but both fuel and water spray are not supplied. For both the conventional and swirl flow structures, the air flow rate is divided into primary and secondary ones, whose ratio varies depending on the combustion load. Table 2.1 shows the air flow rate conditions, where the inflow temperature is set to 40°C in consideration of the compression effect by the blower. The secondary vanes are the guide vanes for the conventional structure, and are swirl vanes with inclination angle of 45° for the swirl flow structure. In addition, the swirl flow structure has a baffle plate as shown in Figure 2.3b, which creates the swirl flow. These numerical simulations of cold flows are solved by FLUENT 17.2 [15]. Each computational domain is divided into approximately 1 million cells. Realizable k - ε model is used as a turbulence model. All walls are non-slip and insulated. Figure 2.5 shows the vertical velocity distribution in the central vertical section at each combustion load obtained by the numerical simulations of cold flows for the large-scale SCV burners with (a) the conventional structure and (b) swirl flow structure. The positive direction is

upwards. (a) In the conventional structure, the flow field in the combustion chamber is biased as the combustion load decreases to 50% and 25%. This is because as the load decreases, the secondary air flow rate unnecessary for combustion increases and is not uniformly distributed. This drift distorts the flame shape and makes it easier for the flame to approach the cold wall, suggesting that overcooling may stop CO oxidation and increase its emissions. (b) The swirl flow structure widens the flow toward the below. The

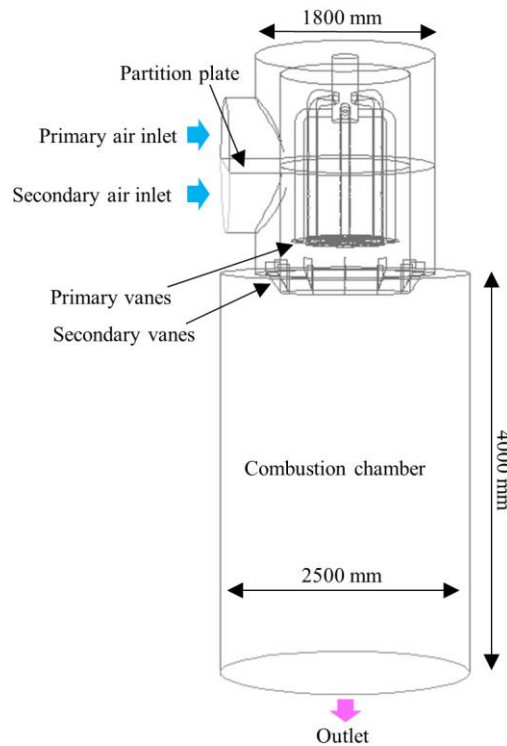


Figure 2.4 Computational domain for numerical simulations of cold flows for large-scale burner with conventional structure.

Table 2.1 Air flow rate conditions for numerical simulations of cold flows. Units are m^3/s at standard state.

Combustion load	Primary air rate	Secondary air rate
100%	12.9	1.43
75%	10.7	3.58
50%	7.15	7.15
25%	3.58	10.7

air drift observed in the conventional structure is no longer confirmed, even at medium and low loads. In addition, the center of the combustion chamber shows negative pressure due to the swirling flow, and a reverse flow region in which the flow is upward is formed. This suggests that the burned gas dilutes the unburned fuel and air and promotes mild combustion, thereby suppressing NO production.

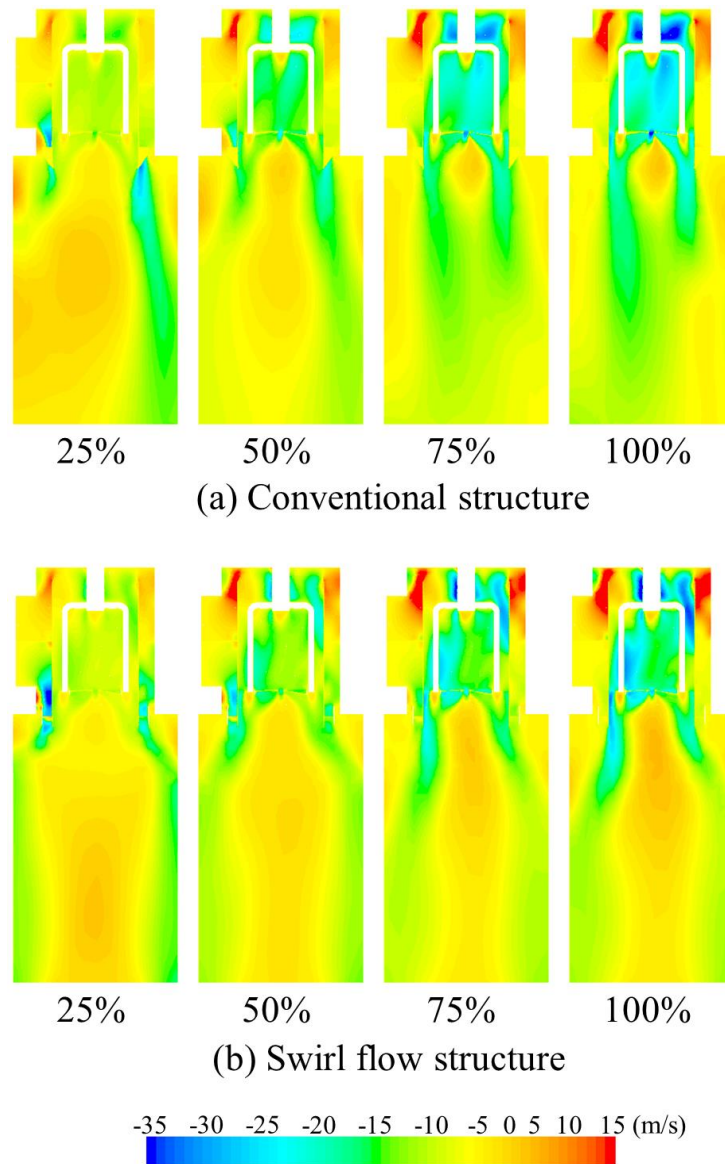


Figure 2.5 Vertical velocity distributions in central vertical section at each combustion load obtained from numerical simulations of cold flows for large-scale SCV burners with (a) conventional structure and (b) swirl flow structure. Positive direction is upwards.

Here, the swirl number S of the swirl flow structure is also addressed, which can be expressed as follows [16]:

$$S = \frac{2}{3} \frac{1 - \left(\frac{D_i}{D_o}\right)^3}{\left[1 - \left(\frac{D_i}{D_o}\right)^2\right]} \tan \alpha, \quad (2.1)$$

where α is the blade inclination angle, D_i is the blade inner diameter, and D_o is the blade outer diameter. For the swirl flow structure, the swirl number of the primary swirl vanes, S_1 , is 0.69 from $\alpha = 45^\circ$, $D_i = 165$ mm, and $D_o = 850$ mm. In addition, the swirl number of the secondary swirl vanes, S_2 , is 0.92 from $\alpha = 45^\circ$, $D_i = 1350$ mm, and $D_o = 1600$ mm. In combustor, the swirl flow induces vortex breakdown and changes the phenomenon of flame propagation, thereby changing the characteristics of blow-off and flashback [17]. The position of the recirculation zone formed by the swirl flow is also related to the occurrence of blowout and flashback [18]. Ito and Song [19] reported that flames under weak swirl conditions, such as $S = 0.13$ or 0.26 , can be easily blown away as compared to those under non-swirl conditions, and the flame stability range is narrowed. On the other hand, under strong swirl conditions such as $S > 0.41$, the flame stability range increases with S . The S for the swirl flow structure can be classified as a strong swirl condition, and this swirl flow is considered effective in stabilizing the flame. As described above, it is predicted that the swirl flow structure weakens the air flow draft, suppresses NO production by forming recirculation zone and improves the flame stability.

Then, the swirl flow structure type large-scale SCV burner with maximum vaporization capacity of 200 t/h has been designed. The developed SCV burner has been installed at Tokyo Gas's Sodegaura LNG Terminal in Fig. 2.6. LESs of water sprayed turbulent combustion are implemented for this burner.



Figure 2.6 Photograph of large-scale SCV burner.

2.2.2 Medium-scale burner

Figure 2.7 shows the schematic of a medium-scale SCV burner. The LNG vaporization capacity is one fiftieth of that of the large-scale SCV burner. The burner has a double pipe structure with an inner pipe diameter of 200 mm and an outer pipe diameter of 320 mm. The fuel flow path is installed in the center and one water spray nozzle is installed at the outer peripheral part in the inner pipe. The water spray nozzle is tilted inward by 20 degrees. Air is supplied from the outer circumference and is distributed to the path through the inner tube and the path through guide vanes. Six guide vanes are installed, and a partition plate is installed on top of them to adjust the pressure loss. The flame formed by the mixture of fuel and air is retained by the burner plate. Figure 2.7b shows the details of the fuel nozzles and the burner plate. There are six fuel nozzles with a diameter of 4.6 mm, and they are inclined 45 degrees toward the outside. The burner plate has twelve holes of 9.8 mm on the inner side and eight holes of 15 mm on the outer side. The burned gas flows out through the combustion chamber with an inner diameter of 440 mm and through a sparge pipe with an inner diameter of 500 mm in the depth direction of the paper. The lower part of the combustion chamber is covered with a pool

of water at room temperature.

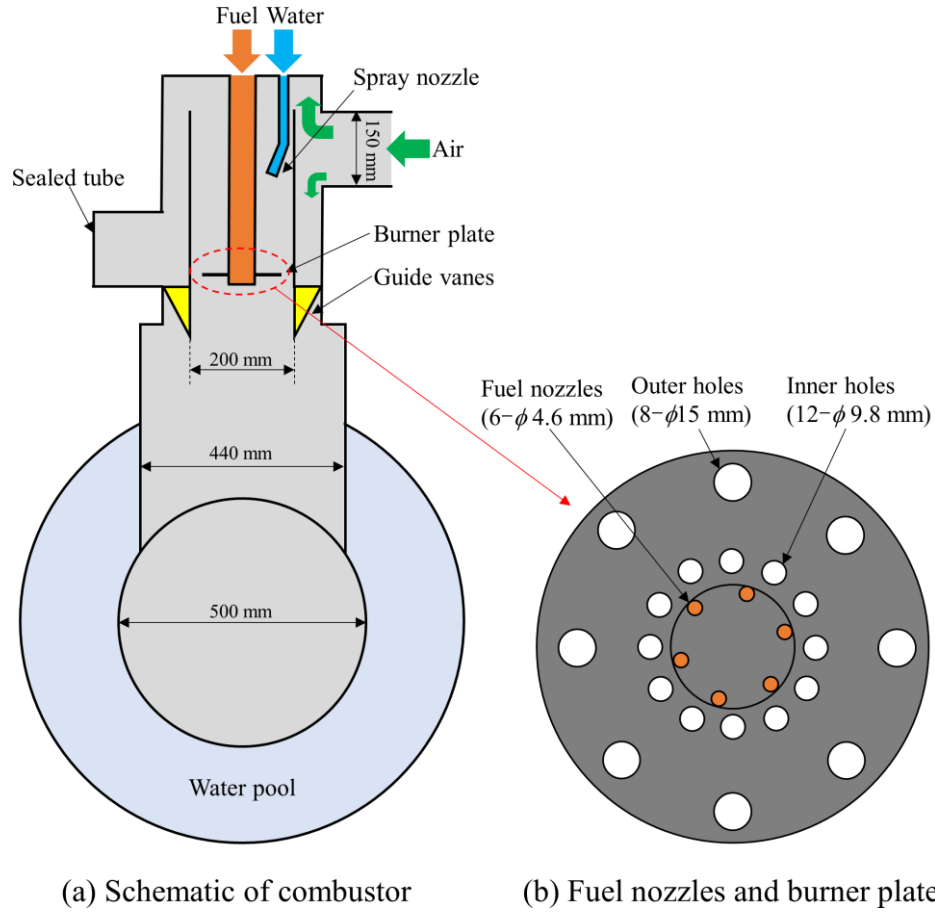


Figure 2.7 Schematic of medium-scale SCV burner. (a) Schematic combustor and (b) Fuel nozzles and burner plate.

2.2.3 Small-scale burner

Figure 2.8 shows a small-scale burner with swirlers manufactured using a 3D printer, which has one spray nozzle mounting port at the center, sixteen fuel nozzles and sixteen vanes with the inclination angle of 45 degrees. The details regarding the spray nozzle is presented in Section 2.4.1. The fuel nozzles' diameter is 1.5 mm. The inner and outer diameters of the swirl vanes are 20 mm and 60 mm, respectively. The swirl number is 0.72 according to the method by Lefebvre et al. [16]. This burner is made of stainless steel 316L. Because the stacking thickness is 20 μ m, the blade thickness of 600 μ m can be sufficiently resolved. Figure 2.9 shows the targeted combustor's (a) appearance and its (b) inside. The combustor is surrounded by insulation castables, and the inner dimensions

are 150 mm $\times$ 150 mm. The burner is attached to the ceiling; fuel, air, and water are injected downward. A heat resistant window is mounted on the front panel to make it possible to observe the inside. The ports for horizontal temperature measurement are attached at heights of 30, 80, 130, 180, and 230 mm below from the burner bottom.

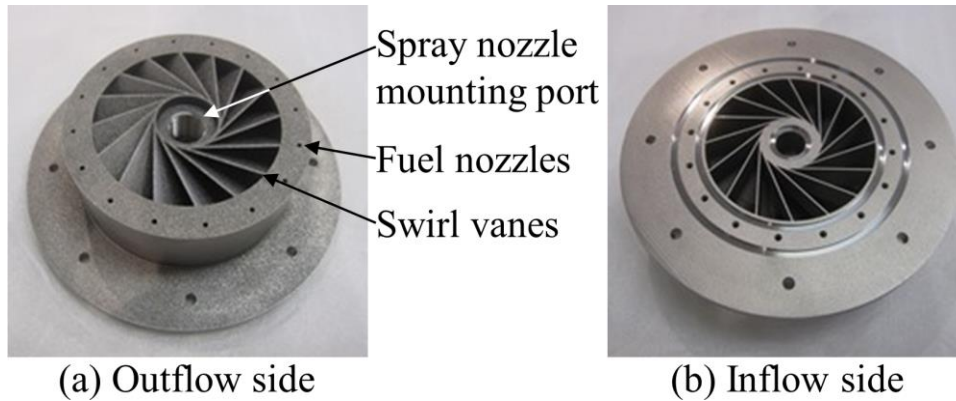


Figure 2.8 Swirl burner manufactured using 3D printer: (a) Outflow side, (b) Inflow side.

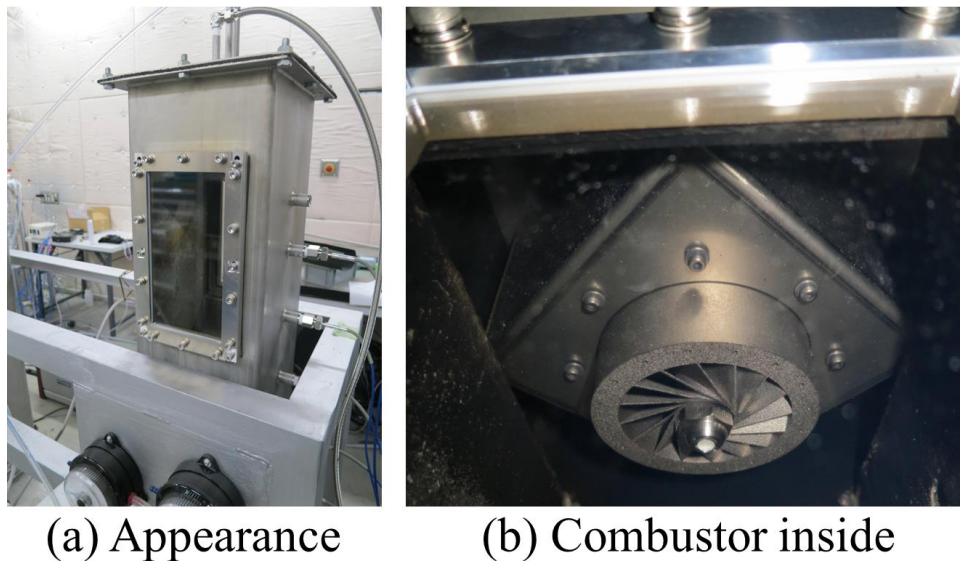


Figure 2.9 Targeted combustor: (a) Appearance, (b) Combustor inside.

2.3 Numerical methods

In this section, the methods of numerical simulations are addressed. First, the 5D-NA-FPV and the 5D-NA-FGM, and LESs employing with them are explained. Th

modeling of water spray and its evaporation is also explained. Next, the neural network modeling is explained. At last, the detail conditions of numerical simulations for each burner.

2.3.1 LES/5D-NA-FPV and LES/5D-NA-FGM

(a) LES/5D-NA-FPV

Non-premixed flamelet libraries based on the conventional FPV [9] are often used for CFD simulations of diffusion combustion. One-dimensional counter diffusion flames are calculated at several scalar dissipation rates for generating the library. However, in a FPV, the set of chemical species' mass fractions in the equilibrium state is basically stored under adiabatic states, and thus, the effect of heat loss on the change in the composition of chemical species cannot be considered. For example, the NO and CO reactions are so slow that they can be easily affected by heat losses. Kishimoto et al. [20] proposed a non-adiabatic procedure that considers the effects of heat losses based on the FPV, which is called non-adiabatic FPV (NA-FPV) approach. For generating the NA-FPV library, the heat release rate is forcibly suppressed during one-dimensional calculations. Thus, absolute enthalpy decreases and the reaction rate and flame temperature decrease. Eventually, the increase in the enthalpy loss can make the flames extinguish.

In addition, in this study, the water vapor mass fraction Z_v is newly introduced for generating the library, to account for the effects of water evaporation and dilution of the fuel and oxidant. Due to the influence of water vapor derived from water evaporation, a mixture of air and water vapor is defined as an oxidant and the water vapor concentration is changed in the one-dimensional counter diffusion flame calculations. Here, the effect of latent heat loss is assumed to be included in the enthalpy loss above. These one-dimensional calculations are performed using a modified version of FlameMaster [21].

The relations among the fuel mass fraction Z , oxidizer mass fraction Z_o , and Z_v is

$$Z + Z_o + Z_v = 1 \quad (2.2)$$

and by assuming that water vapor is part of the oxidant, the mass fraction of water vapor in the oxidant, W , is defined as

$$W = \frac{Z_V}{Z_O + Z_V} = \frac{Z_V}{1 - Z}. \quad (2.3)$$

Physical quantities, φ , such as temperature, density, and mass fraction of chemical species, are represented as

$$\varphi = \varphi(Z, C, \Delta h, W). \quad (2.4)$$

Here, C is the progress variable, which is defined as $C = Y_{CO} + Y_{CO_2} + Y_{H_2} + Y_{H_2O}$, in some researches [22, 23], but as $C = Y_{CO} + Y_{CO_2}$ in this study. This is because Y_{H_2O} is generated through not only the oxidation of fuel but also the evaporation of water droplets, and it is inconvenient for C to increase due to water evaporation. Δh is also enthalpy drop from the adiabatic state. In physical space, the Favre-filtered flamelet library can be obtained as

$$\tilde{\varphi}(\tilde{Z}, \tilde{Z}''^2, \tilde{C}, \tilde{\Delta h}, \tilde{W}) = \int_0^1 \int_0^{\Delta h_{max}} \int_0^1 \int_0^1 \varphi(Z, C, \Delta h, W) \tilde{P}(Z, C, \Delta h, W) dZ dC d\Delta h dW \quad (2.5)$$

where $\tilde{P}(Z, C, \Delta h, W)$ is the density-weighted Favre probability density function and given by following equation:.

$$\tilde{P}(Z, C, \Delta h, W) = B(Z; \tilde{Z}, \tilde{Z}''^2) \delta(C - \tilde{C}) \delta(\Delta h - \tilde{\Delta h}) \delta(W - \tilde{W}). \quad (2.6)$$

Here, δ denotes the delta function and $B(Z; \tilde{Z}, \tilde{Z}''^2)$ is the beta distribution represented as

$$B(Z; \tilde{Z}, \tilde{Z}''^2) = \frac{Z^{\beta_1-1} (1-Z)^{\beta_2-1}}{\int_0^1 Z^{\beta_1-1} (1-Z)^{\beta_2-1} dZ}. \quad (2.7)$$

Here, β_1 and β_2 are given as

$$\beta_1 = \tilde{Z} \left(\tilde{Z} \frac{1 - \tilde{Z}}{\widetilde{Z'^2}} - 1 \right), \quad (2.8)$$

$$\beta_2 = (1 - \tilde{Z}) \left(\tilde{Z} \frac{1 - \tilde{Z}}{\widetilde{Z'^2}} - 1 \right), \quad (2.9)$$

where $\widetilde{Z'^2}$ is the Favre variance of Z . For three-dimensional numerical simulations of combustion fields, referring to the Five-Dimensional NA-FPV (5D-NA-FPV) library expressed by Eq. 2.5, various phenomena such as the chemical reactions, addition of water vapor derived from water evaporation, and heat losses are expressed.

The LESs employing the 5D-NA-FPV (LESs/5D-NA-FPV) are conducted for the large-scale and the medium-scale burners. The governing equations are the filtered conservation equations of mass, momentum, enthalpy $\tilde{h}$, fuel fraction $\tilde{Z}$, water vapor fraction $\widetilde{Z_V}$, progress variable $\tilde{C}$, and NO mass fraction $\widetilde{Y_{NO}}$:

$$\frac{\partial \bar{\rho}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}}) = S_m, \quad (2.10)$$

$$\frac{\partial \bar{\rho} \tilde{\mathbf{u}}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{\mathbf{u}}) = -\nabla \bar{P} + \nabla \cdot \bar{\boldsymbol{\sigma}} + \nabla \cdot \boldsymbol{\tau} + S_u, \quad (2.11)$$

$$\frac{\partial \bar{\rho} \tilde{h}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{h}) = \nabla \cdot (\bar{\rho} \widetilde{D_h} \nabla \tilde{h}) + \nabla \cdot \mathbf{q}_h + Q_{\text{rad}} + S_h, \quad (2.12)$$

$$\frac{\partial \bar{\rho} \tilde{Z}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{Z}) = \nabla \cdot (\bar{\rho} \widetilde{D_Z} \nabla \tilde{Z}) + \nabla \cdot \mathbf{q}_Z, \quad (2.13)$$

$$\frac{\partial \bar{\rho} \widetilde{Z_V}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \widetilde{Z_V}) = \nabla \cdot (\bar{\rho} \widetilde{D_{Z_V}} \nabla \widetilde{Z_V}) + \nabla \cdot \mathbf{q}_{Z_V} + S_{Z_V}, \quad (2.14)$$

$$\frac{\partial \bar{\rho} \tilde{C}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{C}) = \nabla \cdot (\bar{\rho} \widetilde{D_C} \nabla \tilde{C}) + \nabla \cdot \mathbf{q}_C + \bar{\rho} \widetilde{\dot{\omega}_C}, \quad (2.15)$$

$$\frac{\partial \bar{\rho} \widetilde{Y_{NO}}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \widetilde{Y_{NO}}) = \nabla \cdot (\bar{\rho} \widetilde{D_{Y_{NO}}} \nabla \widetilde{Y_{NO}}) + \nabla \cdot \mathbf{q}_{Y_{NO}} + \bar{\rho} \widetilde{\dot{\omega}_{NO}}. \quad (2.16)$$

Here, $\mathbf{u}$ is the velocity, P is the pressure, and $\boldsymbol{\sigma}$ is the stress tensor. D_φ is the diffusion coefficient of each transport scalar φ , which is given as

$$D_\varphi = \frac{\nu}{Pr_\varphi}, \quad (2.17)$$

where ν is dynamic viscosity and Pr_φ is Prandtl number for each φ . $Pr_\varphi = 0.7$ is used for the numerical simulations of combustion fields, τ is the subgrid-scale stress term derived from the turbulence model, and $\mathbf{q}_\varphi$ is the sub-grid scale term. Q_{rad} is the source term of radiative heat transfer. $\bar{\varphi}$ and $\tilde{\varphi}$ denote the LES filtering and Favre averaging of a physical quantity φ , respectively. $\dot{\omega}_C$ is the reaction rate of C . Temperature is calculated from the transported enthalpy with the species' mass fractions and these thermal data.

It is well known that reactions related to NO are slow. The NO concentration in the flamelet libraries is calculated in the equilibrium state. The reactions are often affected by heat losses in practical combustion chambers. In this case, the concentration does not agree with the experimental results. Then, the NO concentration is evaluated based on Ihme and Pitsch's approach [23]. They proposed a method in which the forward and backward NO reaction rates are separately modeled with the reaction rates taken from the flamelet library, as follows:

$$\widetilde{\dot{\omega}_{\text{NO}}} = \widetilde{\dot{\omega}_{\text{NO}}^+} + \tilde{Y}_{\text{NO}} \frac{\widetilde{\dot{\omega}_{\text{NO}}^-}}{\tilde{Y}_{\text{NO}}^{flm}}, \quad (2.18)$$

where $\widetilde{\dot{\omega}_{\text{NO}}^+}$ and $\widetilde{\dot{\omega}_{\text{NO}}^-}$ denote the forward and backward reaction rates, respectively, and $\tilde{Y}_{\text{NO}}^{flm}$ denotes the NO mass fraction in the flamelet libraries. Therefore, for the LES/5D-NA-FPV, the slow rate of the NO reactions under non-adiabatic states can be considered.

(b) LES/5D-NA-FGM

Premixed flamelet libraries based on the conventional FGM [12] are also often used for premixed combustion simulations. One-dimensional free propagation premixed flames are calculated for generating the library. Here, the 5D-NA-FGM is developed based on the FGM through the same procedure as the 5D-NA-FPV.

In physical space, the Favre-filtered flamelet library can be obtained as

$$\tilde{\varphi}(\tilde{Z}, \tilde{C}, \tilde{C}''^2, \tilde{\Delta h}, \tilde{W}) = \int_0^1 \int_0^{\Delta h_{max}} \int_0^1 \int_0^1 \varphi(Z, C, \Delta h, W) \tilde{P}(Z, C, \Delta h, W) dZ dC d\Delta h dW, \quad (2.19)$$

where $\tilde{P}(Z, C, \Delta h, W)$ is given by following equation.

$$\tilde{P}(Z, C, \Delta h, W) = B(C; \tilde{C}, \tilde{C}''^2) \delta(Z - \tilde{Z}) \delta(\Delta h - \tilde{\Delta h}) \delta(W - \tilde{W}). \quad (2.20)$$

Here, δ denotes the delta function and $B(C; \tilde{C}, \tilde{C}''^2)$ is the beta distribution represented as

$$B(C; \tilde{C}, \tilde{C}''^2) = \frac{C^{\beta_1-1} (1-C)^{\beta_2-1}}{\int_0^1 C^{\beta_1-1} (1-C)^{\beta_2-1} dC}. \quad (2.21)$$

Here, β_1 and β_2 are given as

$$\beta_1 = \tilde{C} \left(\tilde{C} \frac{1 - \tilde{C}}{\tilde{C}''^2} - 1 \right), \quad (2.22)$$

$$\beta_2 = (1 - \tilde{C}) \left(\tilde{C} \frac{1 - \tilde{C}}{\tilde{C}''^2} - 1 \right), \quad (2.23)$$

where $\tilde{C}''^2$ is the Favre variance of C . For three-dimensional numerical simulations of combustion fields, the Five-Dimensional NA-FGM (5D-NA-FGM) library expressed by Eq. 2.19 is employed. The governing equations for LES/5D-NA-FGM are same as Eq. 2.10-2.16. The LESs/5D-NA-FGM are implemented for the medium-scale SCV burner. The physical quantities, φ , for each library are simply explained as follows.

$$\varphi = \begin{cases} \varphi(Z, C, Z''^2, \Delta h, W) & \text{for the 5D-NA-FPV} \\ \varphi(Z, C, C''^2, \Delta h, W) & \text{for the 5D-NA-FGM} \end{cases}. \quad (2.24)$$

(c) Water spray and its evaporation

Water spray and its evaporation for the LES/5D-NA-FPV and the LES/5D-NA-FGM is modeled as follows. The water droplets are tracked individually in Lagrangian manner. The phase coupling is implemented using Euler/Lagrangian methods, where source terms (S_m , S_u , S_h , and S_{z_v}) in Eqs. 2.10, 2.11, 2.12 and 2.14 of the governing equations of the gas phase denote source terms due to interaction between gas and water droplet phases and evaluated by the particle-source-in-cell (PSI-Cell) method as follows,

$$S_m = -\frac{1}{\Delta V} \sum_N \frac{dm_d}{dt}, \quad (2.25)$$

$$S_u = -\frac{1}{\Delta V} \sum_N \frac{dm_d \mathbf{u}_d}{dt}, \quad (2.26)$$

$$S_{z_v} = -\frac{1}{\Delta V} \sum_N \frac{dm_d}{dt}, \quad (2.27)$$

$$S_h = -\frac{1}{\Delta V} \sum_N \left[Q_d + \frac{dm_d}{dt} L_v + Q_{rad,d} \right]. \quad (2.28)$$

where ΔV is volumetric size of control volume, N is the number of droplets in a control volume and L_v is latent heat. Q_d and $Q_{rad,d}$ are heats by convection and radiation from gas phase to liquid phase, respectively.

The governing equations for water droplets considering interphase mass, momentum, and heat exchanges can be described as follows:

$$\frac{d\mathbf{x}_d}{dt} = \mathbf{u}_d, \quad (2.29)$$

$$\frac{d\mathbf{u}_d}{dt} = \frac{f_1}{\tau_d} (\tilde{\mathbf{u}} - \mathbf{u}_d), \quad (2.30)$$

$$\frac{dT_d}{dt} = \frac{Nu}{3Pr} \left(\frac{f_2}{\tau_d} \right) \left(\frac{\bar{c}_p}{c_{p,d}} \right) (\tilde{T} - T_d) + \frac{1}{m_d} \left(\frac{dm_d}{dt} \right) \frac{L_v}{c_{p,d}}, \quad (2.31)$$

$$\frac{dm_d}{dt} = -\frac{Sh}{3Sc} \frac{m_d}{\tau_d} \ln(1 + B_M). \quad (2.32)$$

where $\mathbf{x}_d$ is droplet location, $\mathbf{u}_d$ is droplet velocity, T_d is droplet temperature, m_d is

droplet mass. c_p , $c_{p,d}$ are specific heat of gas and droplet respectively, and L_v is the latent heat of the droplet. Nu , Pr , Sh and Sc are Nusselt number, Prandtl number, Sherwood number and Schmidt number, respectively. f_1 and f_2 are correlation coefficients due to the droplet relaxation time. In Eq. 2.30, f_1 is correlation coefficient by the drag force, shown as follows:

$$f_1 = \frac{1 + 0.0545Re_d + 0.1Re_d^{1/2}(1 - 0.03Re_d)}{1 + c|Re_d|^d}, \quad (2.33)$$

$$c = 0.060 + 0.077 \exp(-0.40Re_d), \quad (2.34)$$

$$d = 0.40 + 0.77 \exp(-0.040Re_d), \quad (2.35)$$

where Re_d is the particle Reynolds number. On the other hand, in Eq. 2.31, f_2 is correlation coefficient by the heat transfer given by

$$f_2 = \frac{\beta}{e^\beta - 1}. \quad (2.36)$$

The particle response time of the droplet τ_d in Eqs. 2.33-35 is defined by

$$\tau_d = \frac{\rho_d d_p^2}{(18\mu)}, \quad (2.37)$$

where ρ_d , d_p , and μ are the droplet density, diameter, and viscosity, respectively. B_M is mass transfer number of evaporation, evaluated from mass fractions of the water in the carrier gaseous phase Y_F and at the droplet surface Y_{sf} as follows:

$$B_M = \frac{Y_{sf} - Y_F}{1 - Y_{sf}}. \quad (2.38)$$

Y_{sf} is obtained using the Clausius-Clapeyron relation under saturation pressure condition, as follows:

$$Y_{sf} = \frac{X_{sf}}{X_{sf} + (1 - X_{sf}) \bar{W}/W_F}, \quad (2.39)$$

$$X_{sf} = \frac{P_{atm}}{P} \exp \left\{ \frac{L_V}{R} \left(\frac{1}{T_{BL}} - \frac{1}{T_d} \right) \right\} - \frac{2L_K}{D} \beta, \quad (2.40)$$

where X_{sf} is mole fraction of water vapor at the droplet surface, R is the gas constant for water vapor, P_{atm} is the atmospheric pressure, P is the pressure of carrier gaseous phase. $\bar{W}$ and W_F are the averaged molecular weights of the carrier gaseous phase and the molecular weight of the water vapor, respectively. T_{BL} is the boiling temperature of the liquid droplet. The pressure effect on the boiling temperature is considered as following equations,

$$T_{BL} = \left(\frac{P^{0.119} + b}{11.9} \right)^{\frac{1}{0.119}}, \quad (2.41)$$

$$b = P_{atm}^{0.119} - 11.9 T_{BL,atm}^{0.119}. \quad (2.42)$$

In Eq. 2.40, a non-equilibrium effect is considered using the Langmuir-Knudsen equation law [24-27], and β is non-dimensional evaporation parameter given by

$$\beta = -1.5 Pr \tau_d \frac{1}{m_d} \left(\frac{dm_d}{dt} \right). \quad (2.43)$$

2.3.2 Neural network modeling

To reduce the 5D-NA-FGM library data-size, the employment of deep learning techniques is described. Following two procedures are conducted as the preparation for the neural network modeling. One is the change of the definition of fuel mass fraction Z . In Eq. 2.2, Z means the mass fraction of pure fuel, that is, city gas. Water vapor is contained in $(1 - Z)$, which means it is recognized as part of the oxidant. To generate a neural network, huge amounts of teacher data, which are one-dimensional calculation results, are required. However, there were many cases where the FlameMaster [21] cannot

get the calculation convergence in the definition in Eq. 2.2. This cause is still unknown. The calculation convergence is greatly improved by changing the definition of Z as follows:

$$W = \frac{Z_V}{Z_F + Z_V} = \frac{Z_V}{1 - Z_O} = \frac{Z_V}{Z}. \quad (2.44)$$

That is, water vapor is redefined as a part of the fuel.

Another is the normalization of the progress variable, C . The ranges of Z and W are from zero to one, respectively, and the range of Δh is from zero to the value when the burned gas is at ambient temperature. These ranges are independent of the values of other control variables. On the other hand, the range of C takes changes when the other control variables (Z , W and Δh). Because there are regions of C that does not hold any values, the accuracy of deep learning deteriorates. Then, C is normalized as follows. For the combination of Z , Δh , and W , the maximum value of C , $C_{\max}$, is obtained in advance. The normalized C , C^* , and its variance, $C^{*''}$, are defined as

$$C^* = \frac{C}{C_{\max}}, \quad (2.45)$$

$$C^{*''} = \frac{C}{C_{\max}^2}. \quad (2.46)$$

Therefore, C^* is always within the range from zero to one, and thus, it is not required to learn unnecessary areas, thereby improving learning accuracy. Therefore, the physical quantity ϕ is expressed as $\phi(Z, C^*, C^{*''}, \Delta h, W)$.

A high-dimensional flamelet database, such as the five-dimensional database in this research, requires a large memory size that can exceed the memory capacities of general computers. Therefore, we apply neural networks to express the combustion properties, thereby replacing conventional flamelet tabulation techniques and significantly reducing the required memory size. The regression problem associated with flamelet database is solved using the techniques employed in deep learning. In these neural networks, the input layer is configured according to the control variables of the

flamelet manifold, and the output layer is configured by the physical quantities such as the mass fraction of each chemical species and the production rate of NO_x. The weight parameters of each perceptron are determined using the stochastic gradient descent method. Furthermore, batch renormalization [28] is applied to accelerate the learning process and suppress overlearning. Table 2.2 lists the other methods and the setting parameters. The optimizer and the convergence error is set based on a previous study [13].

The hidden layers of the neural networks were constructed as 11 layers consisting of 9 perceptrons in each layer. However, good convergence solutions could not be obtained using the same configuration network depending on physical quantities owing to the difference in the shape of the supervised data. In particular, this applies to the supervised data with a locally peaky shape, such as radical species, which exists only in a narrow region of the mixture fraction space around the stoichiometric condition. For physical quantities with such properties, the network configuration described above achieved poor convergence. Therefore, the total number of hidden layers is reduced to five, and the number of perceptrons is changed to 9-13-9-5-3. This change is made owing to a theoretical consideration in general regression problems using neural networks, even if there is only one hidden layer, a wide variety of functions can be reproduced if the number of perceptrons is sufficiently abundant. The determination of the network configuration as described above and each method shown in Table 2.2 above are determined by many parametric studies. Furthermore, the depth of the network affects the simulation costs, which requires the calculation of the thermodynamic parameters and the concentrations of chemical species from the function generated in LESs. Therefore, the configuration of the network is comprehensively decided based on the consideration of these factors.

Table 2.2 Methods and setting parameters.

Activation function	$f(x) = \max(x, \tanh(x))$ or ReLU
Optimizer	Adam
Mini batch size	5~10% of all
Epoch number	$1.0 \times 10^4 \sim 3.0 \times 10^4$
Convergence error	2.0×10^{-3}

It takes about several hours to 45 h to calculate each regression, using a GPU node (Tesla P100). Also, the size of data for the flamelet database is compressed from 35 GB to 320 kilobytes (kB).

2.3.3 Computational details

(a) Large-scale burner

Figure 2.10 shows the schematic for a large-scale SCV burner with the swirl flow structure for numerical simulations of combustion fields. The fuel passes through eight supply pipes and is ejected from the fuel injection ports, whose diameter is 7.2 mm, arranged at the tip of each supply pipe. Figure 2.11a shows the calculation grids around the ports.

Air is supplied as the primary and secondary air. Each type of air forms a swirl flow through the baffle plate. Thereafter, the primary air flows downward through the inner cylinder while entraining water droplets ejected from the water spray nozzle. The swirl strength is increased by the primary swirl vanes and flows into the combustion chamber while being mixed with the fuel. The secondary air passes through the secondary swirl vanes provided on the outer periphery and flows to the combustion chamber. The dimensions of the combustion chamber are an inner diameter of 2,500 mm and height of

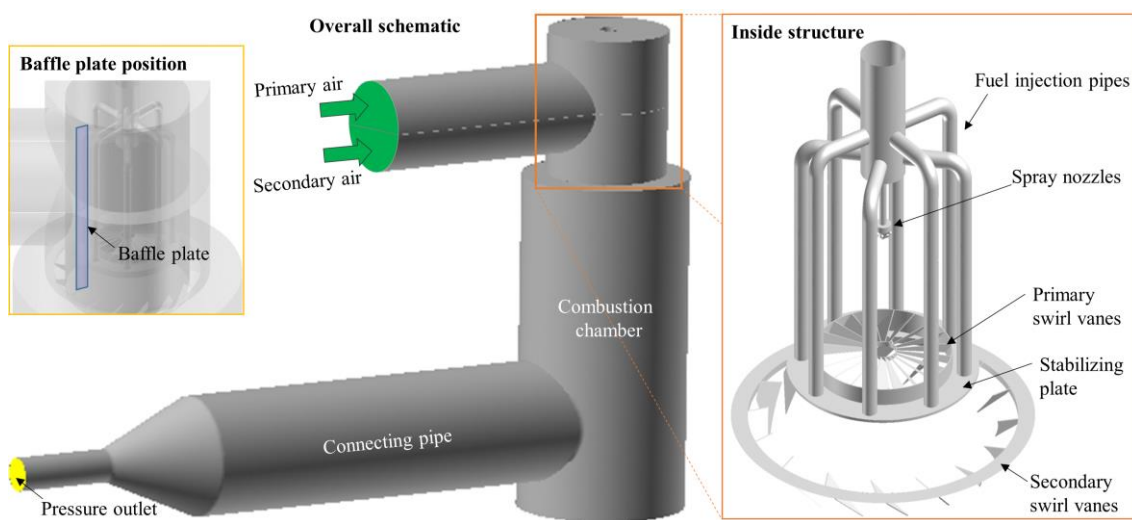


Figure 2.10 Computational domain for numerical simulations of combustion fields for large-scale SCV burner with swirl flow structure.

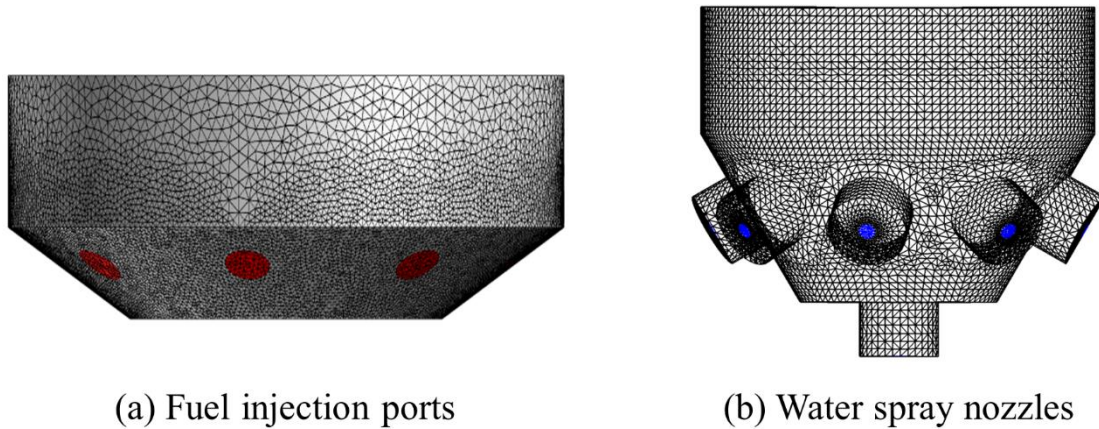


Figure 2.11 Calculation grids for (a) one set of eight fuel injection ports and (b) water spray nozzles with one downward and eight around.

4,500 mm. The burned gas passes through the connecting pipe, with a diameter of 1,800 mm, which connects the combustion chamber and sparge pipes. To reduce the analysis time, the sparge pipe is not simulated and the outlet is narrowed to suppress the back flow from the outside of the calculation region. Figure 2.11b shows the calculation grids near the water spray nozzle. The water spray system has one nozzle facing downward in the center and eight nozzles tilted downward at an intersection angle of 120 degrees. Each spray shape is a hollow cone. The calculation domain faithfully reproduces the actual combustor, and is divided into 541 million cells and 325 million vertices.

The governing equations are solved using an unstructured LES solver: FrontFlow/Red extended by some research institutes, including Kyoto University, referred to as FFR-Comb [7, 10, 20, 29-32]. In this study, the 5D-NA-FPV approach is newly implemented to the code.

In this study, a standard Smagorinsky model is used as the turbulence model and the model parameter C_s is set to 0.1 [33]. A first-order Euler implicit method is used for time advancement, and the time step is set to 2.0×10^{-5} s. The 5D-NA-FPV library is built using the mechanisms developed by the University of California San Diego [34] with nitrogen chemistry [35]. This mechanism has 70 species and 321 reaction steps, and is selected because it has been confirmed that the flame speeds of premixed gas of city gas and air, calculated using the mechanism, are in good agreement with those measured using the heat flux burner [36]. A gray gas model is adopted as the radiation model [37]. The absorption constant is 0.10, which is determined by our experience. The non-

equilibrium Langmuir–Knudsen evaporation model is used as the evaporation model [26, 27, 38].

The boundary conditions are as follows. The fuel temperature is 14°C and the net fuel supply flow rate is 1,700 m³/h in the standard state. The fuel is city gas 13A, and the mixture is set to 91.7% CH₄ and 8.30% C₃H₈ on molar basis. In the standard state, the supply flow rate of primary air is 25,000 m³/h and that of secondary air is 42,000 m³/h. Both air temperatures are 40°C. These conditions correspond to 50% combustion load. The assumed air ratio is 1.4 for the primary air flow rate and 3.7 for the net air flow rate. To investigate the accuracy of NO and CO emissions' prediction under the conditions without/with water spray, the water spray rate condition is set to 0 or 96 L/min. For the condition with water spray, 23 L/min from the central nozzle and 9.1 L/min from each surrounding nozzle are supplied. The spray shape for all nozzles is a hollow cone with a spray angle of 120 degrees. The sprayed water droplet diameters are set to 70 μm for the central nozzle and 60 μm for the surrounding nozzles, based on the experimental measurement of respective nozzles. As the number of water droplets increases in the LES space, the analysis load increases, so 200 droplets are representatively supplied at each time step. Most of the water droplets flow on the primary air flow, but some of them contact the wall surface. The contacted water droplet is completely reflected as an elastic body. As the wall-surface thermal condition, the upper part of the burner is insulated and the wall-surface temperature for the combustion chamber is set to 100°C because the chamber outside is immersed in water.

The scale of the numerical simulations of combustion fields is very large. Therefore, 24,000 parallel calculations are performed using the K computer at RIKEN Center for Computational Science. 3,456,000 CPU hours are required for one case. The actual calculation time per case is approximately 144 hours.

(b) Medium-scale burner

Figure 2.12 shows the schematics of computational domain of the medium-scale burner. The domain is divided into 28 million vertices and 14 million cells. The burner has one air inlet, eight fuel inlets. Their volume flow rates are 2.22×10^{-3} m³/s and 5.00×10^{-2} m³/s in the standard states, respectively. They mix at the downstream side of the burner plate, and a turbulent flame is formed. At the upstream side of that, one water spray

nozzle is installed. The averaged diameter of water droplets is 60 μm , the water flow rate is $3.08 \times 10^{-5} \text{ m}^3/\text{s}$. The jet angle is 90 degrees and the spraying shape is a full-corn type.

The governing equations are solved using the FFR-Comb. The standard Smagorinsky model is used as a turbulence model. The model parameter, C_S , is $C_S = 0.1$. The 5D-NA-FPV or 5D-NA-FGM is used as a turbulent combustion model. The non-equilibrium Langmuir–Knudsen evaporation model is used as the evaporation model. The discrete ordinates model is used for the consideration of the radiative heat transfers [39].

To predict NO and CO emissions for water sprayed turbulent flames, the LES/5D-NA-FPV and the LES/5D-NA-FGM with and without water spray are implemented, respectively. The CPU time required per case of this calculation is approximately 120,000 hours, using the K computer at RIKEN Center for Computational Science. The actual time required for parallel computation of 40,000 steps using 1200 cores is approximately 100 hours of real time.

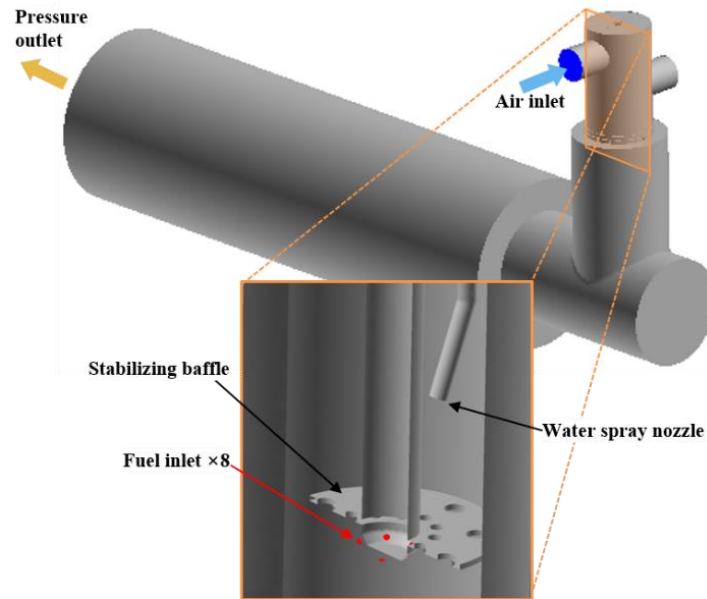


Figure 2.12 Schematics of computational domain of medium-scale burner.

(c) Small-scale burner

Figure 2.13 shows the schematics of computational domain of small-scale burner. The combustor vessel dimension is $150 \text{ mm} \times 150 \text{ mm} \times 1,000 \text{ mm}$, and is narrowed and extended by 355 mm to prevent back flow from outside the analysis region. The domain

is divided into 21.8 million cells. Figure 2.13b shows the internal structure of the swirl burner. The fuel and air inlets are divided into 16 parts, 0.938 L/min and 7.06 L/min under standard state are supplied from each, respectively. The water spray has a flow rate, Q_w , of 22.7 mL/min. The particle size distribution and initial velocity are given based on the Particle Image Analyzer (PIA) measurement results, and the latter has the average value of 2 m/s. The detail of PIA is explained in Section 2.4.1. The heat transfer coefficient of the walls is adjusted according to the experimental results.

The governing equations are solved using the FFR-Comb. The standard Smagorinsky model is used as a turbulence model. The model parameter, C_S , is $C_S = 0.1$. The conventional 5D-NA-FGM or the neural network model is used as a turbulent combustion model. The non-equilibrium Langmuir–Knudsen evaporation model is used as the evaporation model. The discrete ordinates model is used for the consideration of the radiative heat transfers [39].

The LESs are implemented employing either of the following 5D-NA-FGM: the conventional tabulation technique that obtains physical quantities from the library, or the polynomial approximation by the neural network modeling. The time step is set as 50 μ s. Initially 45000 steps are calculated, and then, 25000 steps are calculated to obtain the static values for each parameter.

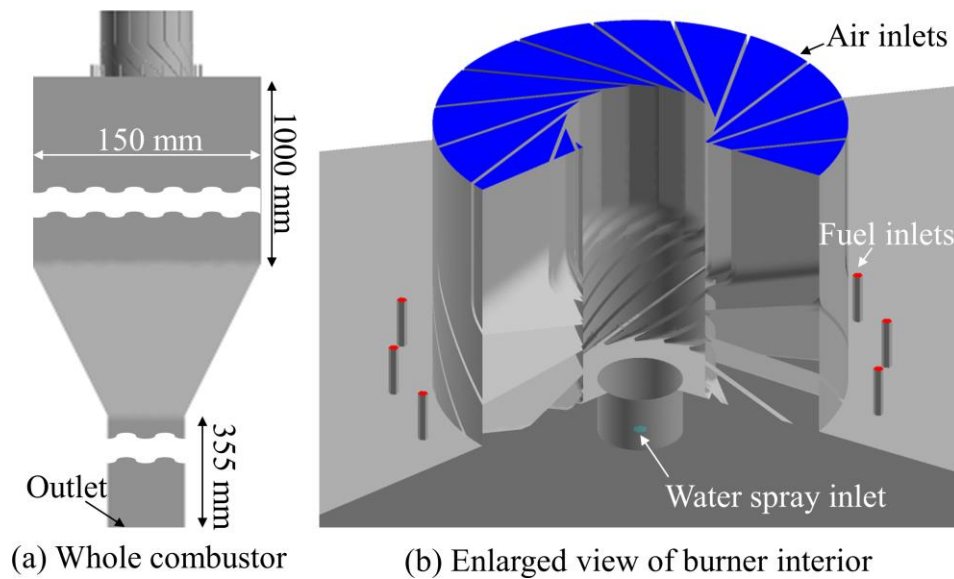


Figure 2.13 Schematics of computational domain of small-scale burner.: (a) Whole region, (b) Enlarged view of burner interior.

2.4 Experimental methods

Combustion experiments using each SCV burner are conducted for comparing the NO and CO emissions with the numerical simulation results. The experimental conditions are explained in this section. First, water spray conditions are explained. Second, the other details are addressed.

2.4.1 Water spray conditions

Because the water spray nozzles installed in the large-scale and medium-scale burners are large, it is difficult to investigate the characteristics in detail. Therefore, the characteristics of the water spray nozzle mounted in the small burner are mainly focused.

(a) Large-scale burner

The large-scale burner has a total of nine water spray nozzles. The layout details are shown in Fig. 2.11b. The spray shape of each nozzle is a hollow cone type. The central nozzle has an average particle size of 70 μm , a spray angle of 120 degrees and a flow rate of 23 L/min, and the peripheral nozzles have an average particle size of 60 μm , a spray angle of 120 degrees and a flow rate of 9.1 L/min.

(b) Medium-scale burner

The spray shape of the nozzle installed in the medium-scale burner is a full cone type, and the average particle size is 60 μm , the spray angle is 90 degrees, and the flow rate is 1.85 L/min. The full cone type is introduced not to hinder uniform spray by the burner plate in the burner.

(c) Small-scale burner

The spray characteristics of the water spray nozzle mounted in the small-scale burner is investigated in detail. The spray shape is a hollow cone type. A stable water spray is supplied from a water pool pressurized using compressed air. The distributions of the particle size and flow velocity required for numerical simulations are measured using a PIA employing a double pulse Nd:YAG laser and a charge-coupled device (CCD)

camera [40]. The CCD has an effective length of 8.8 mm and width of 6.6 mm, and a pixel size of $5.5 \mu\text{m} \times 5.5 \mu\text{m}$. The water spray nozzle faces downward. The measurement is carried out while shifting by 50 mm below the bottom in the radial direction with $r = 0.00, 12.5, 18.8, 25.5$ mm. The distributions are obtained by changing the spray pressure. Figure 2.14 shows the particle size distributions at each position, at gauge pressure of 0.3 MPa. The highest flow rate is observed at approximately $r = 18.8$ mm, and the average particle size is $57.7 \mu\text{m}$. The water droplets measured at $r = 0.00$ mm are very fine, and the flow rate is negligible.

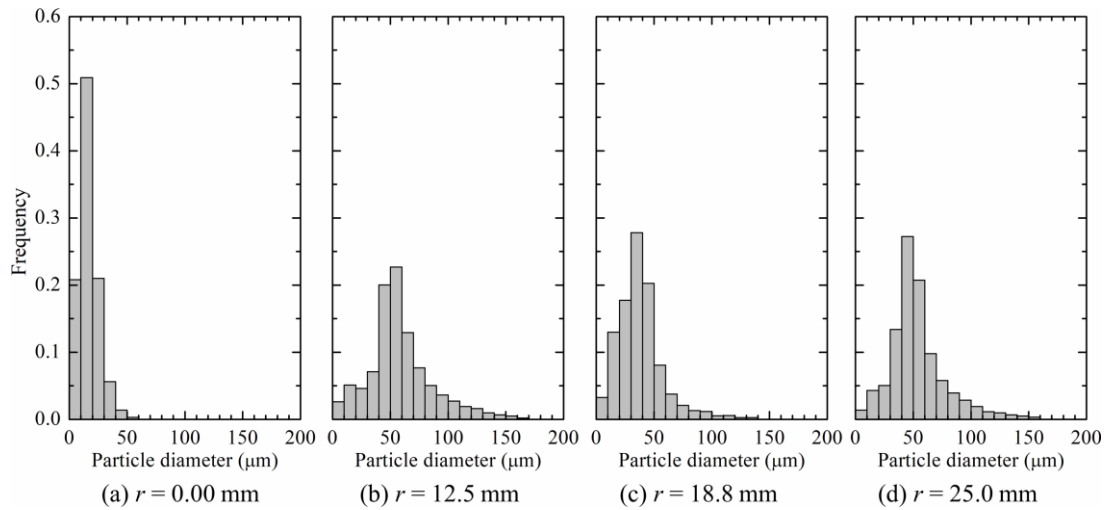


Figure 2.14 Particle size distributions at each position: (a) $r = 0.00$ mm, (b) $r = 12.5$ mm, (c) $r = 18.8$ mm, (d) $r = 25.0$ mm.

2.4.2 Experimental procedure

For all burners, the fuel is city gas 13A, whose higher heating value (HHV) is 45 MJ/m^3 at standard state, and the oxidant is ambient air. And combustion emissions are measured using an exhaust gas analyzer of constant potential electrolytic type at the flue outlet. NO, nitrogen dioxide (NO_2), CO, O_2 and CO_2 concentrations are measured, respectively. Other experimental procedures for each burner is described as follows.

(a) Large-scale burner

Combustion experiments for a large-scale SCV burner with the swirl flow structure are conducted. Figure 2.15 shows the direct photograph inside the large-scale SCV burner,

which is taken from a peep window attached to the burner top. Although the flame shape cannot be clearly recognized, a stable combustion is confirmed under the primary swirl vanes. The experiments start with 100% combustion load condition. NO and CO emissions at various water spray amounts are measured while reducing the combustion load. The flow rates of the fuel and air are estimated from each differential pressure. The water spray system is installed inside, and the maximum water spray flow rate is about 100 L/min.

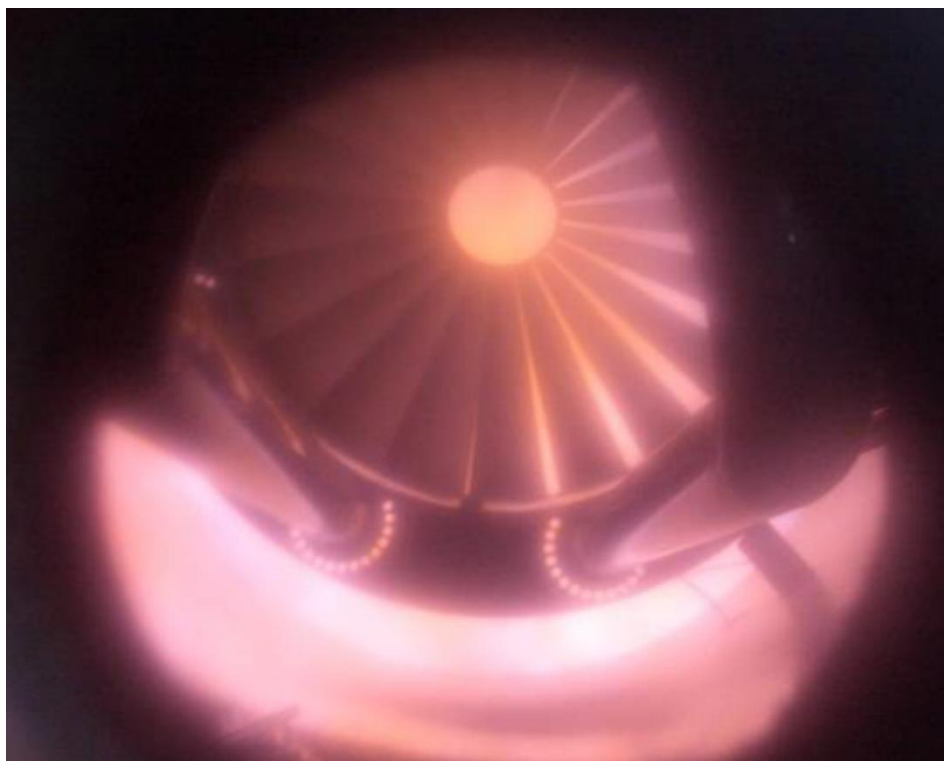


Figure 2.15 Direct photograph inside large-scale SCV burner.

(b) Medium-scale burner

The flow rates of the fuel and air are controlled by each mass flow meter. The combustion experiments using the medium-scale burner aim to verify the prediction accuracy of the NO and CO emissions of the LES/5D-NA-FPV and LES/5D-NA-FGM. Therefore, the combustion load and the water spray amount are not changed to focus on two conditions with and without water spray.

(b) Small-scale burner

The flow rates of the fuel and air are controlled by each mass flow controllers. A mixture composed of 10 L/min of the fuel and 5 L/min of air is supplied from the fuel nozzle, and 113 L/min of air is supplied through the swirl vanes. The total excess air ratio is 1.10. The fuel-rich mixture is supplied instead of pure fuel to prevent the generation of soot. Temperature and combustion emissions are measured as follows. The temperature distributions are measured using R-type thermocouples, which are composed of Pt-Pt/13%Rh and have a wire diameter of 100 μm . To suppress the catalytic reaction on the thermocouples in the presence of flames, the surface of the thermocouples is coated by broiling hexamethyldisiloxane [41]. The combustion emissions are measured at the flue of the combustor, using an exhaust gas analyzer of controlled potential electrolysis sensors or galvanic cell sensors. The measurement error for combustion emissions is 5%.

2.5 Results and discussion

2.5.1 Water spray effects

The water spray effects on combustion fields are described for each burner.

(a) Large-scale burner

First, the NO and CO emissions when the water spray amount is constant are examined. Because if the water spray is stopped at high loads, the burner is damaged by the overheating, the results at 65 L/min are focused on here. Figure 2.16 shows the measurement results of NO and CO emissions at various combustion loads for the large-scale SCV burner with the swirl flow structure when the water spray amount is fixed at 65 L/min. NO concentrations are corrected at $\text{O}_2 = 5\%$ and CO concentrations are raw values. The NO emissions slightly increase with the combustion load, however, are below 50 ppm in the whole region. The swirl flow structure has an upwardly convex emission characteristic about CO emissions, exceeding 110 ppm at 50% load. Figure 2.17 shows the measurement results of NO and CO emissions under various water spray rate conditions at each load of (a) 100%, (b) 50%, and (c) 25% for the large-scale SCV burner. NO concentrations are corrected at $\text{O}_2 = 5\%$ and CO concentrations are raw values. NO emissions can be reduced less than 50 ppm by using water spray. On the other hand, CO

is hardly detected at 100% load and is emitted at 60 ppm or more even without the water spray at loads of 50% and 25%. CO emissions increase at medium and low loads because the secondary air flow rate increases and the flow hits the side wall of the combustion chamber, due to the swirling flow by the secondary swirl vanes, as shown in Fig. 2.5. It

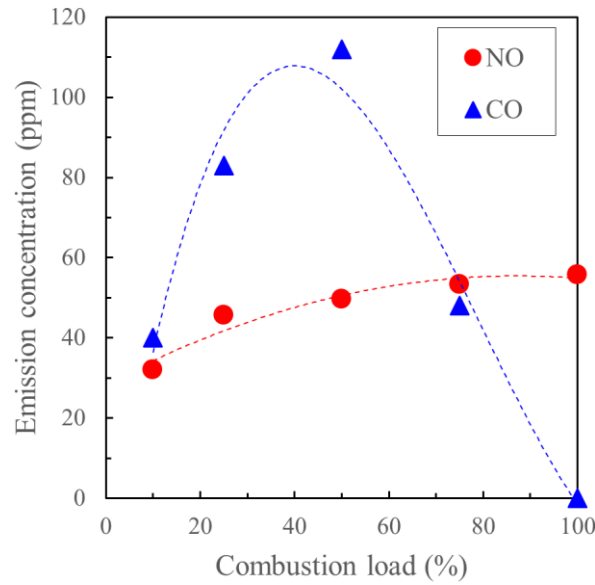


Figure 2.16 NO and CO emissions measured at various combustion loads for large-scale SCV burner with swirl flow structure. Water spray rate is 65 L/min. NO concentrations are corrected at $O_2 = 5\%$ and CO concentrations are raw values.

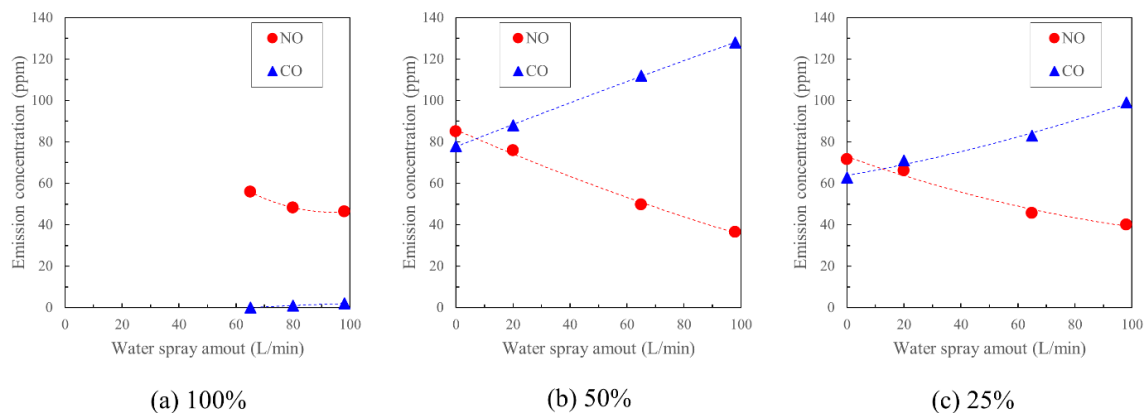


Figure 2.17 NO and CO emissions measured in various water spray rate conditions at each load of (a) 100%, (b) 50% and (c) 25% for large-scale SCV burner with swirl flow structure. NO concentrations are corrected at $O_2 = 5\%$ and CO concentrations are raw values.

is thought that CO oxidation is suppressed by contacting the cold wall. At the 50% load, the NO and CO emissions are 85 ppm and 78 ppm under the condition without water spray. When water spray amount is 98 L/min, they are 37 ppm and 128 ppm, respectively.

From the combustion experiments of the large-scale SCV burner in the previous section, it is confirmed that NO and CO emissions are large at especially 50% load. Here, finally, the results of numerical simulations of combustion fields for the large-scale SCV burner with the swirl flow structure are discussed. To confirm whether NO and CO emissions without/with water spray condition can be predicted using the 5D-NA-FPV approach, which can take into account the heat loss of water evaporation, numerical simulations of combustion fields are conducted in two cases, where each water spray condition is 0 and 98 L/min.

The effect of water spray on the combustion state is examined. Figure 2.18 shows the 1400°C temperature iso-surface in red and the water droplets in blue particles under the conditions (a) without water spray and (b) with water spray. The swirling flow of air forms a complex turbulent flow field, as well as a flame surface, while mixing with fuel.

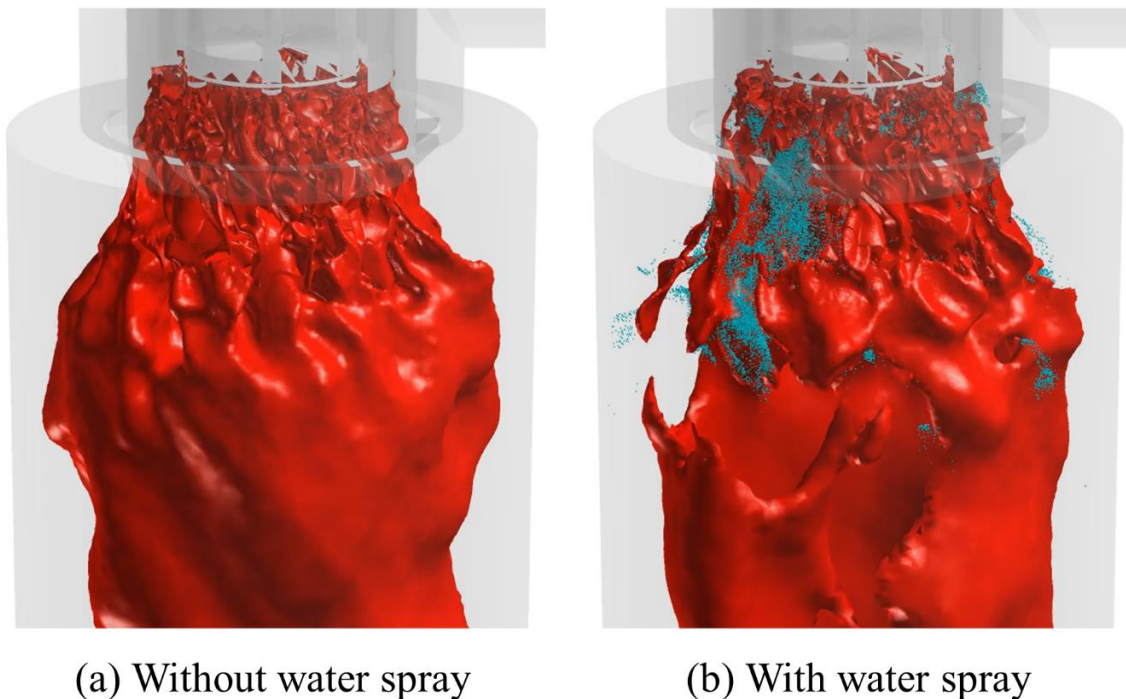


Figure 2.18 Instantaneous results of 1400°C temperature iso-surface in red and water droplets in blue particles under conditions of (a) without water spray and (b) with water spray.

In the condition without water spray, a cylindrical flame is formed and no extinguishing point is observed. In the condition with water spray, the flame surface partially disappears due to the latent heat of evaporation of the water droplets and water droplets that have not completely vaporized pass through the flame surface. In addition, the injected water droplets are almost completely evaporated in the upper space of the combustion chamber.

Figure 2.19 shows the time-averaged distributions of H_2O mole fraction under the conditions of (a) without water spray and (b) with water spray. In the condition with water spray, to obtain the H_2O mole fraction, the water vapor generated by the water droplets' evaporation is added to that generated by the fuel combustion. The sum is shown as Net H_2O in Fig.2.19b, where the H_2O concentration in the center of the combustion chamber increases due to evaporation. From the above, it is confirmed that the water spraying and water evaporation phenomena can be expressed using the 5D-NA-FPV approach.

Figures 2.20-2.22 show the distributions of speed, temperature, and CH_4 mole fraction obtained by time averaging in the vertical center cross-section, respectively. In Fig. 2.20, the flow field hardly changes, irrespective of the presence or absence of water

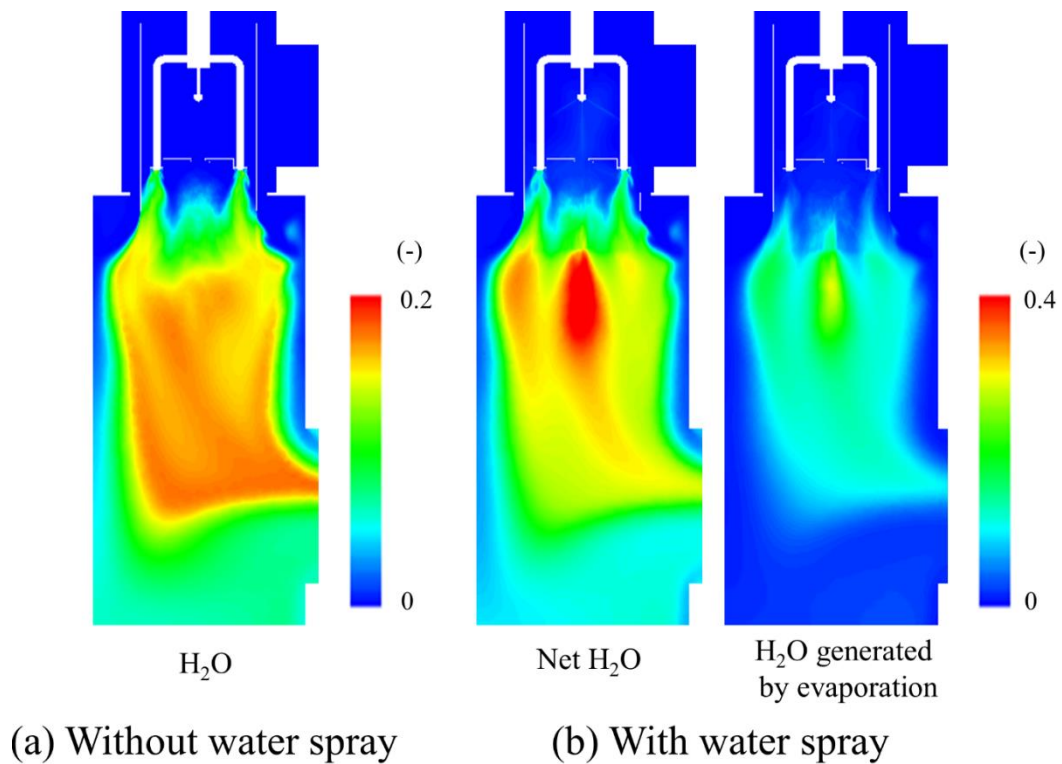


Figure 2.19 Time-averaged distributions of H_2O mole fraction under conditions of (a) without water spray and (b) with water spray.

spray. The two swirl vanes strengthen the flow in the outer peripheral portion and stagnate it in the central portion of a backflow region. In Fig. 2.21, the temperature in the combustion chamber in the condition with water spray is reduced by about 500°C,

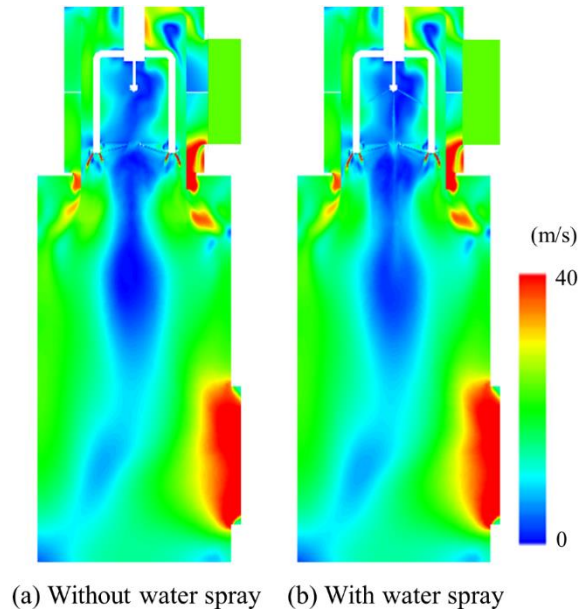


Figure 2.20 Time-averaged distributions of speed obtained under conditions of (a) without water spray and (b) with water spray.

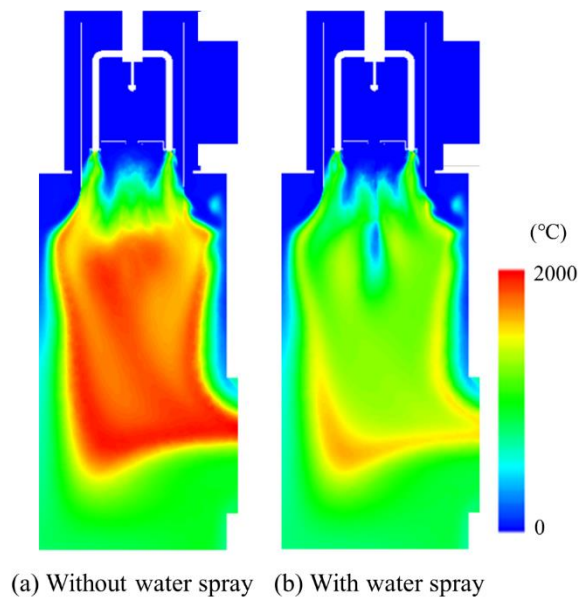


Figure 2.21 Time-averaged distributions of temperature obtained under conditions of (a) without water spray and (b) with water spray.

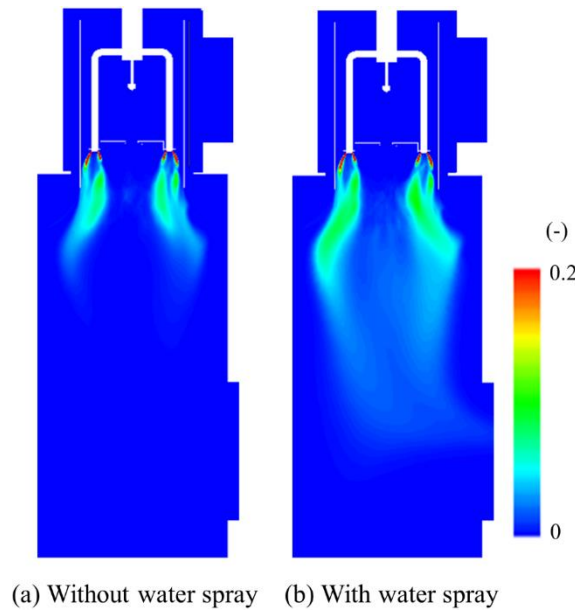


Figure 2.22 Time-averaged distributions of CH_4 mole fraction obtained under conditions of (a) without water spray and (b) with water spray.

compared to that in the condition without water spray. The temperature drop region corresponds to the region where the vapor concentration derived from evaporation is high, as shown in Fig. 2.19b. Most water droplets are evaporated in the upper space of the combustion chamber, and a high-temperature region is generated again in the lower space. A rapid temperature drop leads to a decrease in reaction rate. Focusing on the CH_4 mole fraction distributions in Fig. 2.22, CH_4 is completely consumed in the upper part in the condition without water spray, whereas a part of CH_4 in the condition with water spray continues to burn near the exit of the combustion chamber. In this way, it is possible to capture how the combustion reaction is slowed down by the latent heat loss of water evaporation.

Figure 2.23 and 2.24 show the NO mass and the CO mole fraction distributions in the vertical center section in the conditions (a) without water spray and (b) with water spray. The NO concentration is determined by solving a transport equation that takes into account the production and consumption rates of NO. Figure 2.23b shows that NO production in the center of the combustion chamber is suppressed in the condition with water spray. NO concentrations in Fig. 2.23a and 2.23b around the bottom do not change

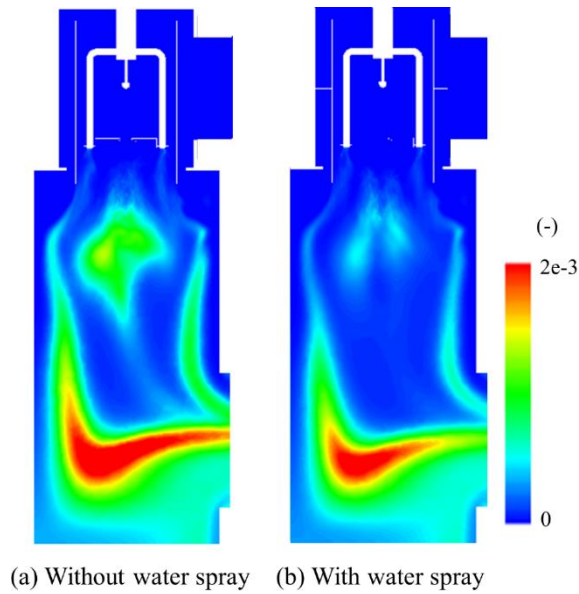


Figure 2.23 Time-averaged distributions of NO mass fraction obtained under conditions of (a) without water spray and (b) with water spray.

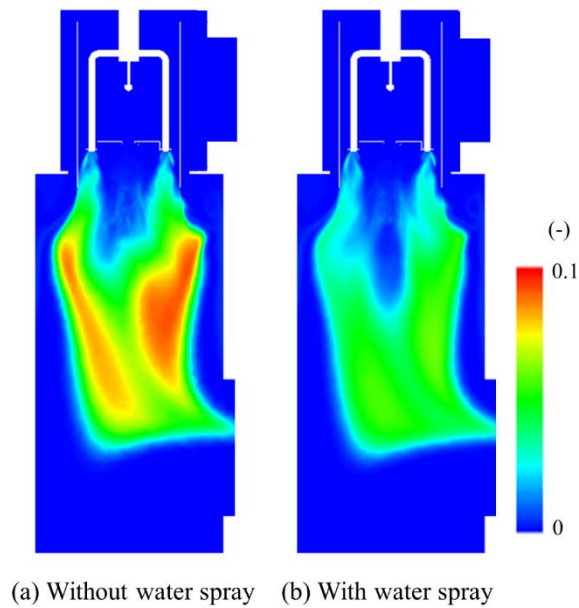


Figure 2.24 Time-averaged distributions of CO mole fraction obtained under conditions of (a) without water spray and (b) with water spray.

much. This region, where the NO concentration is high, coincides qualitatively with the region where the temperature is high in Fig. 2.21. In the condition without water spray, shown in Fig. 2.24a, there is a cylindrical region where the CO concentration is high along the wall of the combustion chamber. This can be understood as the fact that CO is

produced from CH_4 , by looking at the region where CH_4 is consumed in Fig. 2.22a. On the other hand, in the condition with water spray, shown in Fig. 2.24b, a remarkable high-concentration region is not formed. As shown in Fig. 2.22b, as the oxidation rate of CH_4 is slow, this behavior is reasonable.

(b) Medium-scale burner

The effect of the heat loss by water evaporation is investigated. As an example, Fig. 2.25 shows the instantaneous distributions of (a) temperature and (b) W by the LES/5D-NA-FGM. The region where W increases corresponds that where temperature decreases. This indicates the flame is cooled by latent heats of vaporization. Figure 2.26 shows time-averaged temperature distributions by (a) the LES/5D-NA-FGM or (b) the LES/5D-NA-FPV in conditions with and without water spray. Both of flames by the LES/5D-NA-FGM and the LES/5D-NA-FPV without water spray are attached the burner plate. The flame length for the LES/5D-NA-FGM is longer than that for the LES/5D-NA-FPV. Also, the maximum flame temperature for the LES/5D-NA-FPV becomes slightly higher. The

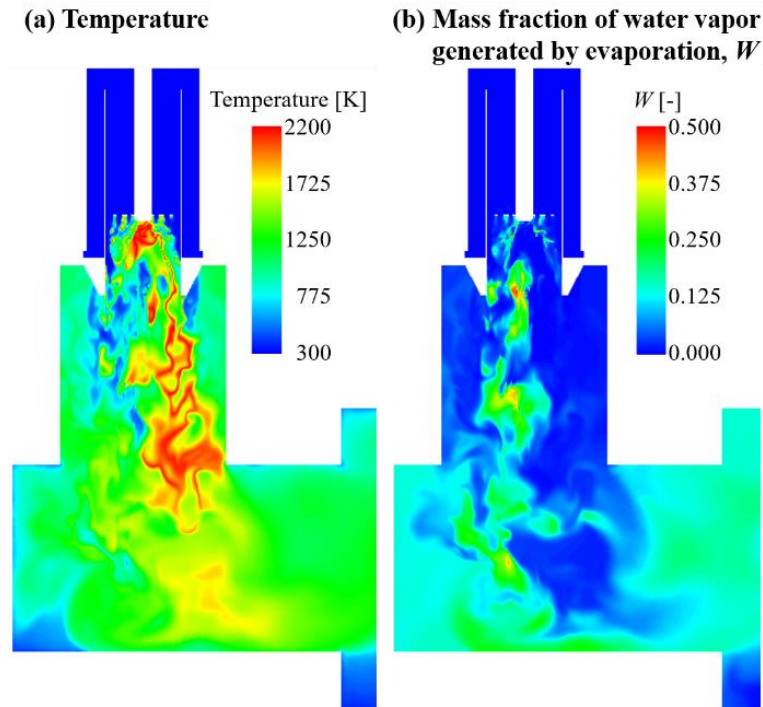


Figure 2.25 Instantaneous distributions by LES/5D-NA-FGM in condition with water spray: (a) temperature, (b) W .

water spray causes the maximum flame temperature to drop by more than 400 K. Figure 2.27 shows time-averaged distributions of W by (a) the LES/5D-NA-FGM or (b) the LES/5D-NA-FPV. Because the flame for the LES/5D-NA-FPV has short length and high temperature, it is reasonable that the water evaporation rate for the LES/5D-NA-FPV is totally faster. The W is high on the left side in Fig. 2.25b, but its time-averaged distribution is almost symmetrical in Fig. 2.27a. This indicates that the region where the evaporation is active is time-varying. As above, both the LES/5D-NA-FGM and the LES/5D-NA-FPV

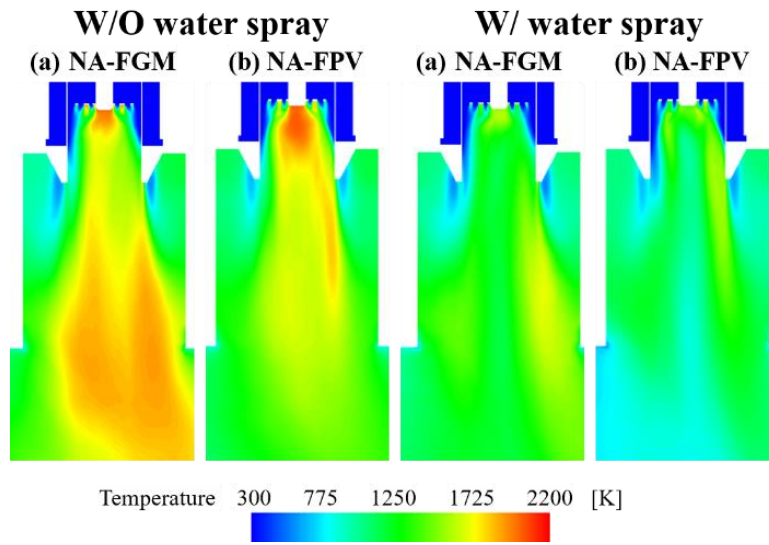


Figure 2.26 Time-averaged distributions of temperature by (a) LES/5D-NA-FGM and (b) LES/5D-NA-FPV in conditions with and without water spray.

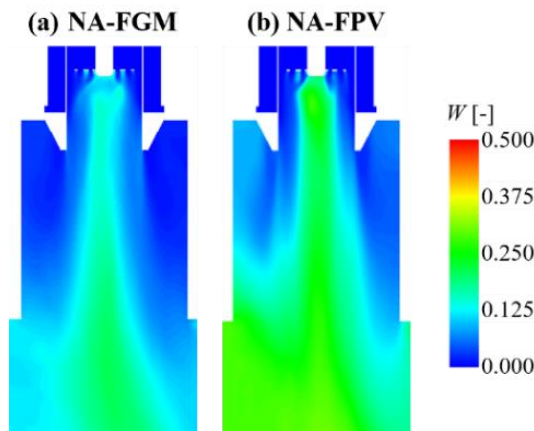


Figure 2.27 Time-averaged distributions of mass fraction of water vapor generated by evaporation, W , by (a) LES/NA-FGM and (b) LES/NA-FPV.

can consider the heat losses, but the characteristic of each flame is quite different.

Figure 2.28 shows time-averaged distributions of CO mole fraction by (a) the LES/5D-NA-FGM and (b) the LES/5D-NA-FPV in conditions with and without water spray. The consumption rate of CO for the LES/5D-NA-FGM is slower than that for the LES/5D-NA-FPV. In the region of high W in Fig. 2.27a, the CO concentration become high in the condition with water spray. This indicates that the consumption of CO is

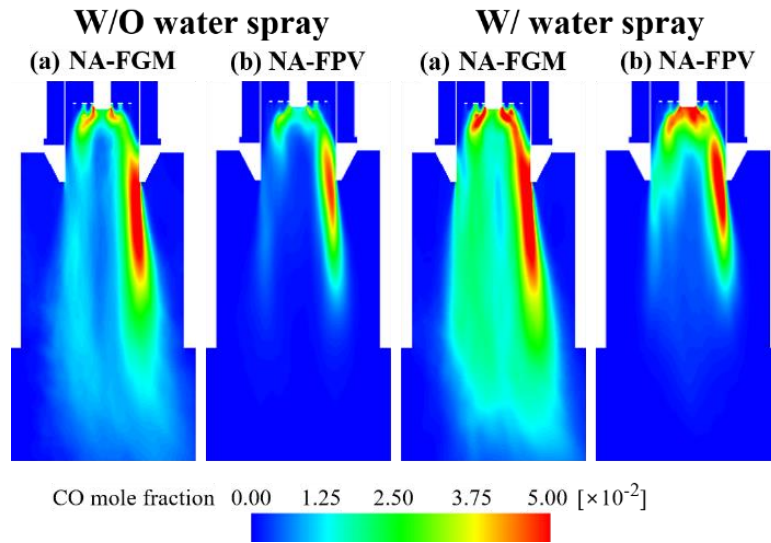


Figure 2.28 Time-averaged distributions of CO mole fraction by (a) LES/NA-FGM and (b) LES/NA-FPV in conditions with and without water spray.

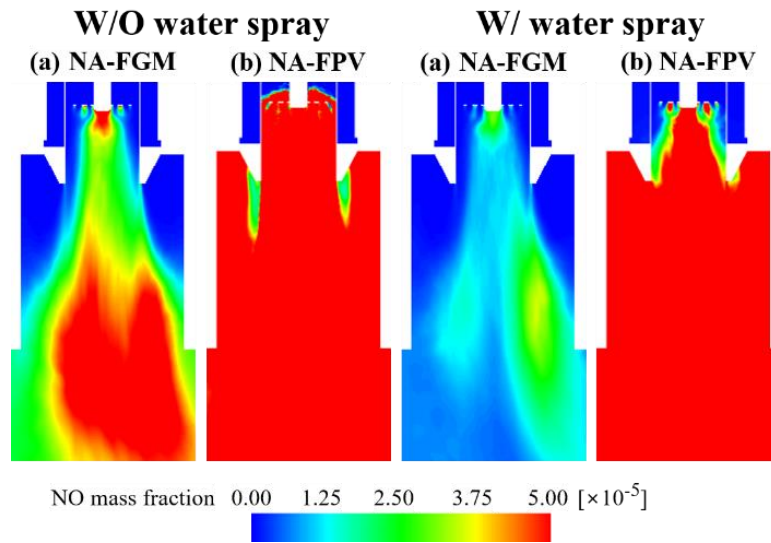


Figure 2.29 Time-averaged distributions of NO mass fraction by (a) LES/NA-FGM and (b) LES/NA-FPV in conditions with and without water spray.

suppressed by water's evaporations. Figure 2.29 shows time-averaged distributions of NO mass fraction by (a) the LES/5D-NA-FGM and (b) the LES/5D-NA-FPV in conditions with and without water spray. For the LES/5D-NA-FGM, the regions of high NO concentrations correspond with high temperature regions in Fig. 2.26. NO concentrations for the LES/5D-NA-FPV is quite higher than them for the LES/5D-NA-FGM.

(c) Small-scale burner

Figure 2.30 shows the images of turbulent flames in two conditions, i.e., no water spray and $Q_w = 22.7$ mL/min. Each flame is conical, and the flame base is attached to the bottom of the swirl vanes. High brightness is observed below the fuel nozzles. The unburned fuel moves in the backflow generated by the swirl flow is burned around the bottom of the swirl vanes. In Fig. 2.30b, it can be seen that the water vapor generated by the evaporation is heated and thus emits red light.

Figure 2.31 shows the NO, NO₂, and CO emissions measured at the flue with respect to the water spray flow rate. Each concentration is presented as a dry basis and is corrected at 0% O₂. As the flow rate increases, the NO and NO₂ emissions decrease and increase, respectively. At the flow rate of 22.7 mL/min, the NO₂ emissions exceed NO emissions. The CO emissions increase with increasing water spray flow rate and exceed 2000 ppm at the flow rate of 22.7 mL/min. The CO emissions cannot be measured at the

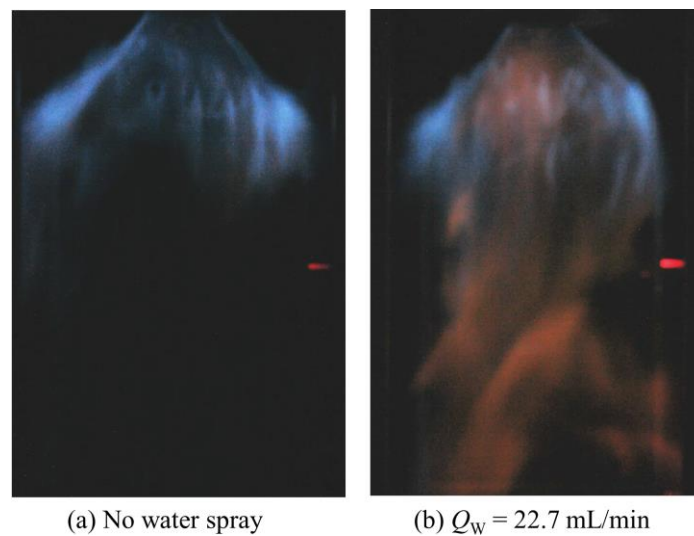


Figure 2.30 Images of turbulent flames: (a) no water spray and (b) $Q_w = 22.7$ mL/min.

flow rate of 28.7 mL/min due to exceeding 10,000 ppm, which is beyond the measurable limit of the apparatus. In addition, the flame at the flow rate of 33.3 mL/min becomes unstable and eventually extinguishes.

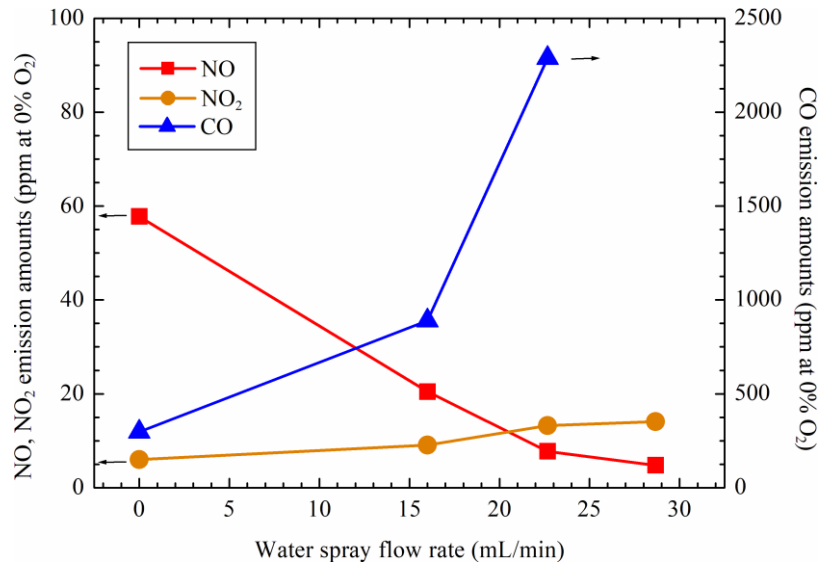


Figure 2.31 Measurement results of NO, NO₂ and CO emission amounts against water spray flow rate.

2.5.2 Predictions of NO and CO emissions

The predictions of NO and CO emissions by the LESs/5D-NA-FPV and the LESs/5D-NA-FGM are discussed for the large-scale and the medium-scale burners. Those for the small-scale burner are discussed in the next section.

(a) Large-scale burner

Finally, Fig. 2.32 shows the comparisons of NO and CO emissions between the simulation and experimental results under the conditions of with and without water spray. The NO concentrations are corrected at O₂ = 5% and the CO concentrations are raw values. The horizontal axis indicates the water-spray flow rate, and the vertical axis indicates each concentration. The NO emissions predicted by the numerical simulations of combustion fields are overestimated by 2 to 3 times compared to the experimental values. The tendency of NO emissions to decrease with increasing water spray flow rate is well captured. In addition, the CO emissions predicted by the numerical simulations of

combustion fields are underestimated by about 2.3 times. However, this also captures the tendency of CO emissions to increase with the water spray flow rate.

It is found that the behaviors of NO and CO emissions affected by water spray can be evaluated by LES/5D-NA-FPV, which will be used for the optimal design of practical burners.

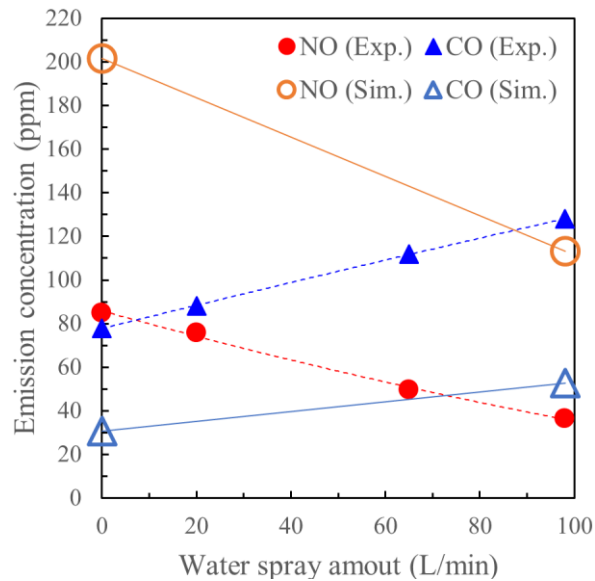


Figure 2.32 Comparison of NO and CO emissions between simulation and experimental results in conditions with and without water spray. NO concentrations are corrected at $O_2 = 5\%$ and CO concentrations are raw values.

(b) Medium-scale burner

Figures 2.33 and 2.34 show the change of NO and CO emissions with and without water spray among the calculations by the LES/5D-NA-FGM and the LES/5D-NA-FPV and the experiments. The experimental measurement values are obtained by using an exhaust gas analyzer of constant potential electrolytic type at the flue outlet of the SCV burner. First, CO emissions are focused on. For the experiments, water spray multiplies CO emissions from 48 ppm to 328 ppm. The LES/5D-NA-FGM slightly overestimates CO emissions as compared with the experiments, and they are predicted as 88 ppm for the condition without water spray and 434 ppm with water spray. On the other hand, the LES/5D-NA-FPV underestimates CO emissions massively. Second, NO emissions are focused on. All NO emissions are corrected by $4\%O_2$ in Fig. 2.33. Water spray for the

experiments decreases NO emissions from 58 ppm to 28 ppm. The LES/5D-NA-FGM predicts the close values to the experiments. However, the LES/5D-NA-FPV

overestimates NO emissions by around ten times in comparison with the experiments and the LES/5D-NA-FGM.

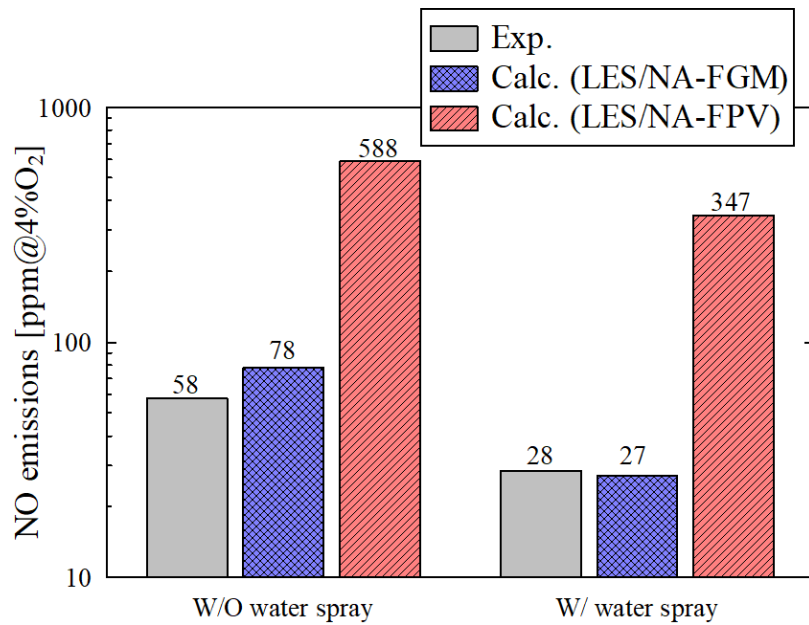


Figure 2.33 Comparison of NO emissions among calculations by LES/NA-FGM and LES/NA-FPV in conditions with and without water spray and experiments.

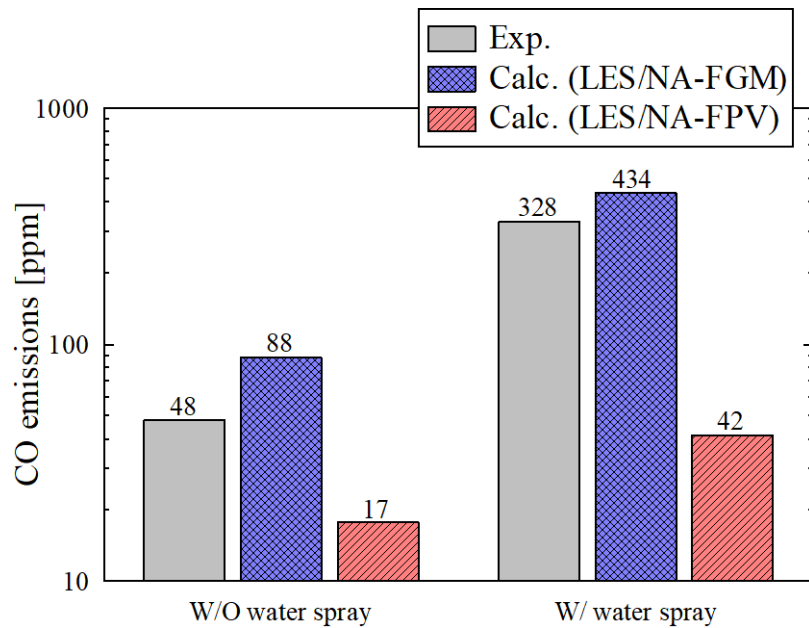


Figure 2.34 Comparison of CO emissions among calculations by LES/NA-FGM and LES/NA-FPV in conditions with and without water spray and experiments.

As observed above, both of the LES/5D-NA-FGM and the LES/5D-NA-FPV can express the increase of CO emissions and the decrease of NO emissions by water's evaporations. In this study, NO and CO emissions predicted by the LES/5D-NA-FGM agree well with the experiments much better than the LES/5D-NA-FPV. It is examined the reason why the LES/5D-NA-FGM excels at the predictions of NO and CO emissions. Figure 2.35 shows streamlines around the burner plate. Lots of large vortexes are generated at the downstream of the plate, which makes the fuel and the air mix well. It is presumable that the flame of the SCV burner primarily has the characteristic of premixed combustion by forming premixed turbulent flames after the mixture.

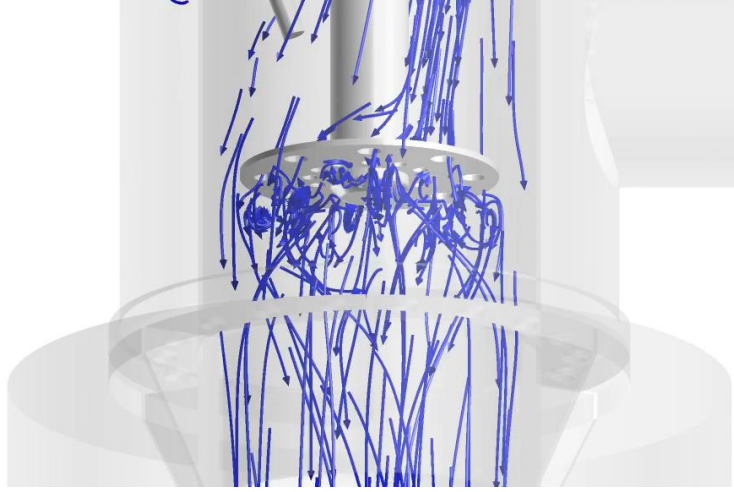


Figure 2.35 Streamlines around burner plate.

2.5.3 Validation of neural network modeling

Figure 2.36 shows (a) temperature, (b) reaction rate of C , $\dot{\omega}_C$, (c) CO mass fraction, (d) production rate of NO, and (e) consumption rate of NO with respect to the normalized progress variable, C^* , predicted using the conventional database and the neural network model, respectively. The results for $W = 0, 0.2$, and 0.4 are plotted. Here, $Z = 0.0512$, $C^{*''} = 0.00$, and $\Delta h = 0.00$ are fixed. The temperature and $\dot{\omega}_C$ can be accurately predicted. For the CO mass fraction and the production/consumption rates of NO, the errors increase when $C^* > 0.6$, but they are generally good agreement with those in the database.

Figure 2.37 shows the comparison of the time-averaged temperature distributions among experiments and simulations employing the database or the neural network model

in the conditions (a) without water spray and (b) with water spray. The temperature trends for both simulation results employing the neural network model and the conventional database are similar to the experiment shown in Fig. 2.37a. To predict the lower temperature distributions for simulation results in whole is an issue. There may be problems in the setting of the wall boundary conditions or radiation models. The results of the LES employing the neural network model are good agreement with those employing the conventional technique. In Fig. 2.37b, the temperature of the simulation

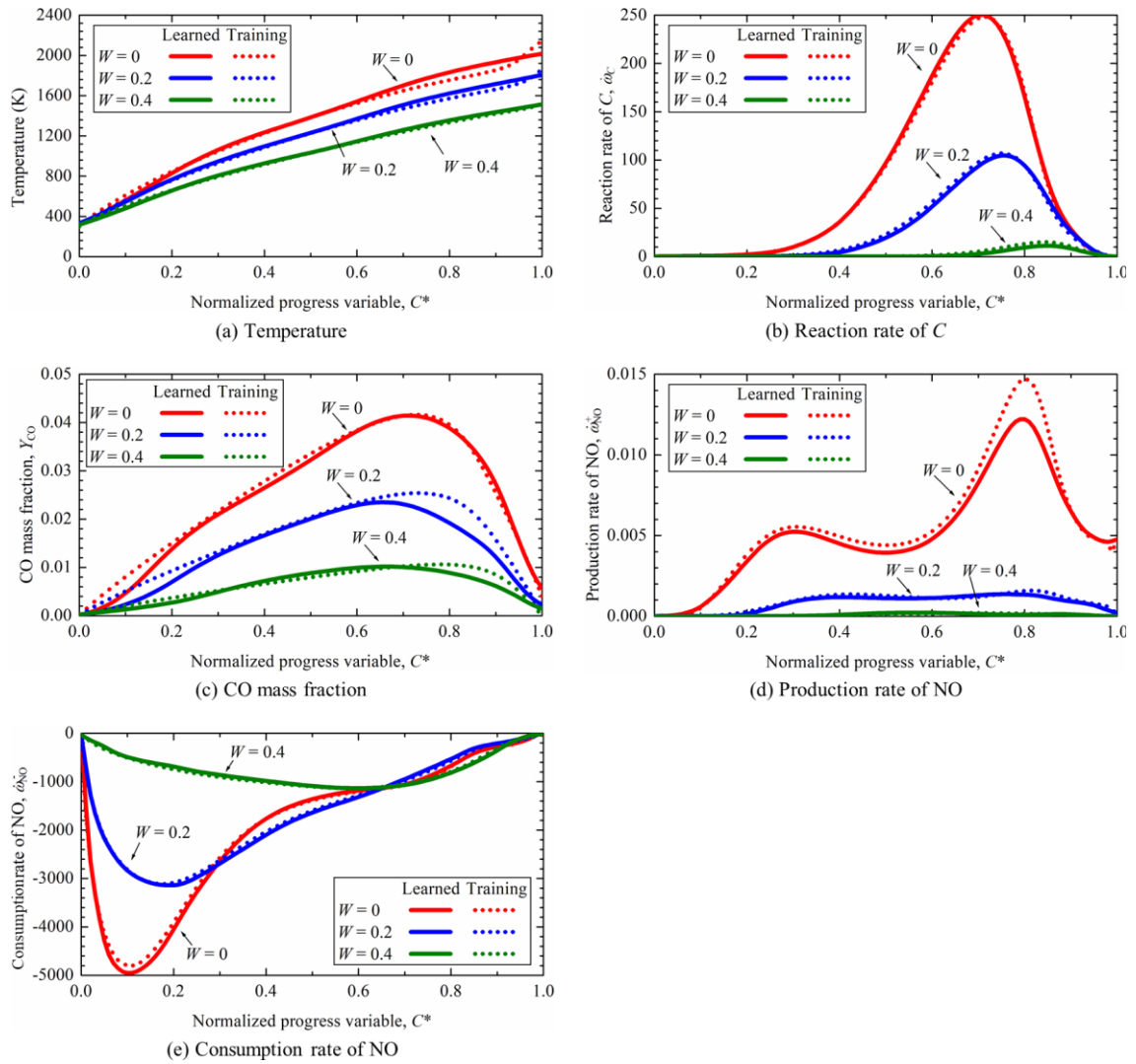


Figure 2.36 (a) Temperature, (b) Reaction rate of C, ω_C , (c) CO mass fraction, (d) Production rate of NO, and (e) Consumption rate of NO with respect to normalized progress variable, C^* , predicted using conventional database and neural network model for $W = 0, 0.2, \text{ and } 0.4$.

employing the neural network model decreases compared to the condition without water spray. For the quantitative evaluation, further verification is necessary for the considerations of the water evaporation.

For the LESs employing the neural network model, Figs. 2.38-2.40 show the time-averaged distributions of the mass fractions of water vapor, CO, and NO in the conditions (a) without water spray and (b) with water spray, respectively. In Fig. 2.38b, the region where water vapor is generated is slightly downstream from the region of Fig. 2.30b. This indicates that the evaporation rate in the simulation is slow or the backflow effect accompanying swirl flow is weak. Moreover, CO and NO mass fractions increase and decrease due to the cooling effect of the water spray, respectively. These behaviors are similar to the experimental results presented in Fig. 2.31. From the above mentioned discussion, the effects of the water spray on the combustion state can be generally expressed through the LESs employing the neural network model.

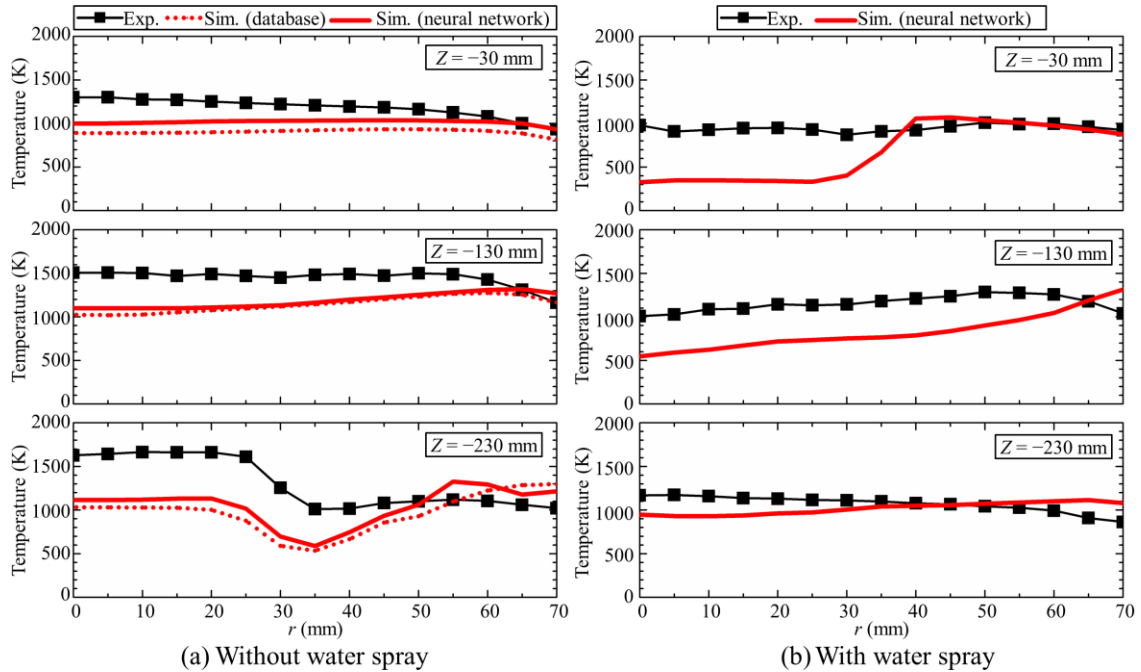


Figure 2.37 Time-averaged temperature distributions among experiment and simulation employing database or neural network model: (a) Without water spray and (b) With water spray.

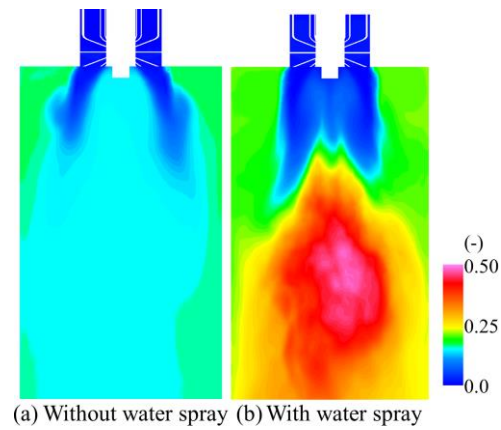


Figure 2.38 Time-averaged distributions of water vapor mass fraction: (a) Without water spray and (b) With water spray.

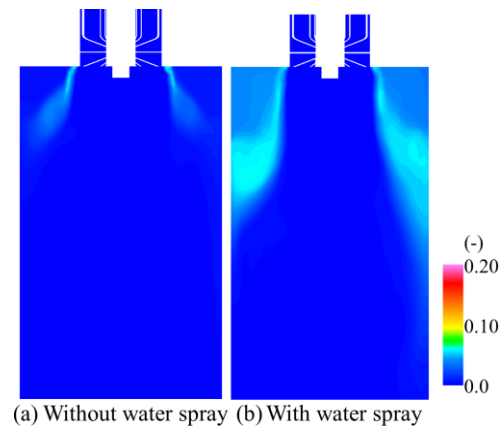


Figure 2.39 Time-averaged distributions of CO mass fraction: (a) Without water spray and (b) With water spray.

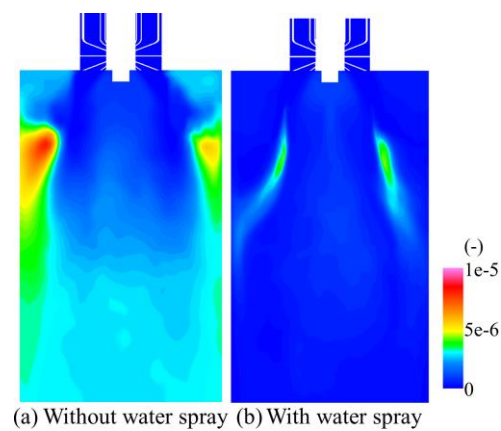


Figure 2.40 Time-averaged distributions of NO mass fraction: (a) Without water spray and (b) With water spray.

2.6 Conclusions

In this chapter, turbulent flames in SCV burners equipped with water spray system were investigated through the LESs and the experiments. Three types of SCV burners with different sizes were considered. The large-scale SCV burner was developed by numerical simulations of cold flows, and the characteristics of NO and CO emissions were investigated by the LESs/5D-NA-FPV and the experiments. For the medium-scale SCV burner, the validities of NO and CO emissions by the LESs/5D-NA-FGM and the LESs/5D-NA-FPV was compared. For the small-scale SCV burner, the neural network model was developed based on the 5D-NA-FGM. The main results with respect to three purposes in this chapter are summarized as follows.

Characteristics of NO_x and CO emissions and validity of LESs/5D-NA-FPV for a large-scale SCV burner:

- (1) For the large-scale SCV burner, the NO emissions decrease as the water spray amount increase and record less than 50 ppm corrected at O₂ = 5% regardless of the loads, whereas the CO emissions increase as the water spray amount increase and reach 120 ppm at approximately 50% load.
- (2) The combustion fields generated using the large-scale SCV burner are generally reproduced by LESs/5D-NA-FPV. The LESs/5D-NA-FPV can account for the latent heat loss, which lowers the flame temperature and suppresses the combustion reactions. Furthermore, it can qualitatively capture the trends of the effects of the water spray flow rate on the NO and CO emissions, that is, they decrease and increase with increasing water spray rate, respectively. This suggests that it can be a powerful tool for the optimal design of large-scale SCV burners.

Validities of LES/5D-NA-FGM and LES/5D-NA-FPV for a medium-scale SCV burner:

- (1) Both LESs/5D-NA-FGM and LESs/5D-NA-FPV can account for the heat losses due to water evaporation and the suppression of NO production and CO consumption.
- (2) The NO and CO emissions predicted by LESs/5D-NA-FGM agree much better with the experiment results than those of LESs/5D-NA-FPV in the medium-scale SCV

burner. This is because the fuel and air are mixed well around the burner plate, and the characteristics of premixed flames become evident.

Use of a neural network model based of 5D-NA-FGM for a small-scale SCV burner:

- (1) The CO concentrations and reaction rates of NO as well as the temperature and production rates of the progress variable predicted by the neural network model are in good agreement with those of the flamelet database of the training data, but further improvements are required to accurately predict the CO and NO concentrations with local peaks for the control variables.
- (2) The temperature trends for both simulation results using the neural network model and the conventional technique are generally captured in the experiment in the absence of water spray. In the condition with water spray, the water evaporation caused the temperature decrease, CO increase, and NO production suppression, as in the experiment. For a quantitative evaluation, it is necessary to improve the accuracy of the neural network model and boundary conditions after examining the validity of the flamelet method.

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Chapter 3: Ammonia/methane premixed swirl combustion

3.1 Introduction

The emissions of carbon dioxide (CO_2), which is thought to cause global warming, is increasing at a rapid rate. In Japan, 39% of CO_2 emissions are produced in energy transformation industries, such as power generation plants [1]. The amount of CO_2 emissions in gas power plants is approximately one half of that in coal-fueled power plants [2]. The demand for gas power plants is expected to increase, but to reduce CO_2 emissions will remain as an urgent issue.

Ammonia (NH_3) combustion has recently drawn intense research interest because of its potential to reduce CO_2 emissions when implemented in such power plants. Ammonia is 17.8% by weight hydrogen, a relatively high hydrogen ratio [3, 4]. The technologies related to its production, transportation, and storage are well established, with ammonia being the second largest produced chemical in the world. Ammonia can be also produced from renewable energy sources [5]. In addition, ammonia can be easily liquefied and stored in the conditions of 8.5 bar at ambient temperature or 240 K at ambient pressure, which is less expensive than liquefying pure hydrogen [6]. Therefore, ammonia has been considered as a potential source of hydrogen and an alternative carbon-free fuel for the future.

Considerable research on ammonia combustion has been done since the 1960s [7-9]. Experimental studies showed that the minimum ignition energy of ammonia is 16 times higher than that of fossil fuels. At stoichiometric conditions, the quenching distance of ammonia/air was 3.5 times greater than that of propane/air [8]. Other experiments demonstrated that ammonia/air has a relatively slow chemical reaction rate, giving a laminar burning velocity of 6-8 cm/s [10], which is only one fifth that of methane/air. In other studies, high production of nitrogen oxides (NO_x) originating from nitrogen in NH_3 is considered as a problem [11-13].

In the face of such problems for ammonia combustion, some challenges for the practical application have been conducted recently. Valera-Medina et al. [14, 15]

combusted ammonia/methane/air in various equivalence ratios in a laboratory-scale tangential swirl burner and suggested low swirl and different injection strategies to optimize gas turbines with ammonia as a primary fuel. Kurata et al. [16] demonstrated ammonia/methane/air combustion employing a micro gas turbine in the Fukushima Research Energy Institute. Hayakawa et al. [17] showed that a specific equivalence ratio supports low emissions of nitric oxide (NO) and unburned ammonia.

Because NO_x emissions are strictly regulated, an optimized burner design for ammonia combustion is required. For such purposes, computational fluid dynamics (CFD) can be an effective approach. Somarathne et al. [18] illuminated the effect of secondary air injection to reduce the amount of NO emissions. It was suggested that the equivalence ratio, ϕ , at the primary zone should be controlled as $\phi = 1.2$. This indicates that the equivalence ratio in an ideal combustion chamber changes from a fuel-rich condition to a stoichiometric condition. Additionally, the NO production rate is relatively slow and depends on temperature. Therefore, to evaluate the amount of NO emissions, it is necessary to consider heat losses in combustion chambers. However, the simulations in previous researches were conducted in an adiabatic state using detailed reaction mechanisms [18, 19]. There is no study that examined the validity of turbulence combustion models, especially flamelet approaches.

The purpose of this chapter is, therefore, to perform large-eddy simulation (LES) using two types of flamelet approach for ammonia/methane/air premixed combustion in the swirl burner [14], and to validate our method by comparing the amounts of NO and carbon monoxide (CO) emissions with experimental data. At the beginning, in order to investigate an adequate mechanism for the condition of the swirl burner, some representative detailed reaction mechanisms are compared through evaluating the laminar burning velocity and the amount of NO emissions. Then, LESs with two types of flamelet approach are conducted. As a combustion model, a Non-Adiabatic Flamelet Generated Manifold approach (NA-FGM), which is extended based on the conventional flamelet-generated manifold approach (FGM) [20]. The NA-FGM can consider the effects of various heat losses through radiation and cold walls. Proch and Kempf [21] employed the NA-FGM for a tabulated premixed flame. In their approach, product mass fraction, product mass fraction variance and enthalpy difference were introduced for generating the flamelet library, whereas mixture fraction is additionally introduced in this study, in

order to take into account the variation of ϕ . In addition, the NO and CO reactions are discussed in non-adiabatic states from the viewpoint of chemical reactions

3.2 Numerical methods

3.2.1 Targeted chamber and swirl burner

Figure 3.1 shows schematics of computational domain and generic swirl burner. The detailed structure of this burner is identified in the previous studies [14, 15]. The fuel consists of 61.0% NH_3 and 39.0% CH_4 by volume, and the oxidizer is air. The ammonia/methane/air premixed gas is supplied from the inlet nozzle, and, after passing through the swirler, it is burned in the combustion chamber made of a quartz glass. The domain is divided into ~ 5 million vertexes and ~ 6 million cells.

As boundary conditions, the volume flow rate of fuel at the inlet is constant as $1.35 \times 10^{-3} \text{ m}^3/\text{s}$. The inlet premixed gas temperature is 298 K, and the ambient pressure is 1 atm. If the LES/NA-FGM is employed, temperature, convection heat transfer rate, and emissivity on the walls of the combustion chamber are set to 1,500 K, $10 \text{ W/m}^2\text{K}$, and 0.8 respectively. The other walls are treated adiabatically. On the other hand, the all walls are treated adiabatically if the conventional LES/FGM is employed.

Here, the swirl number, Sw , of the burner is also addressed. Sw can be defined as

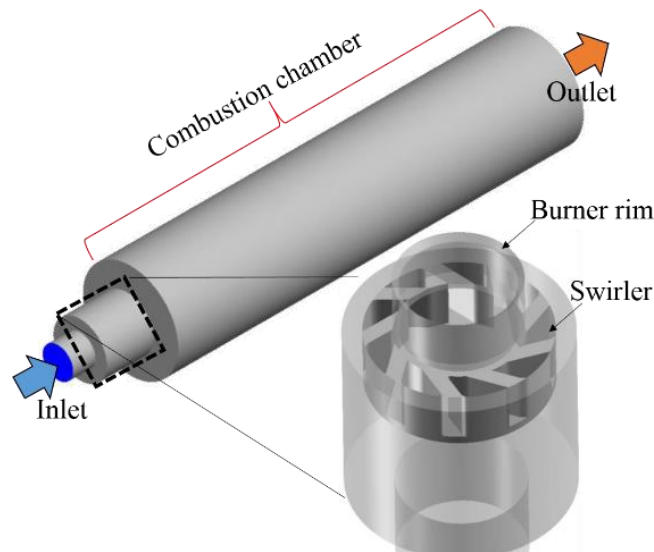


Figure 3.1 Schematics of computational domain and generic swirl burner.

$$Sw = \frac{G_a}{G_r r_1}, \quad (3.1)$$

where G_a and G_r are respectively the angular and radial momenta, and r_1 is the radius of the tubular cavity of the swirler. Velocity at the slit, w_s , is expressed as

$$w_s = \frac{Q}{n_s t_s L_s}, \quad (3.2)$$

where Q is total volume flow rate, and n_s , t_s and L_s are respectively the number of slits, the thickness and the longitudinal length for each slit. The angular momentum per unit volume for the premixed gas is $\rho w_s (r_1 - t_s/2)$. Here, $(r_1 - t_s/2)$ is the distance between the axis and the center of the slits. Because of the above, the total angular momentum, G_a , is expressed as

$$G_a = \rho Q w_s \left(r_1 - \frac{t_s}{2} \right) = \rho Q^2 \frac{r_1 - \frac{t_s}{2}}{n_s t_s L_s}. \quad (3.3)$$

On the other hand, mean axial velocity, U , is expressed as

$$U = \frac{Q}{\pi r_1^2}. \quad (3.4)$$

Accordingly, the total radial momentum, G_r , is expressed as

$$G_r = \rho Q U = \frac{\rho Q^2}{\pi r_1^2}. \quad (3.5)$$

Therefore, Sw is expressed as

$$Sw = \frac{1}{n} \frac{\left(r_1 - \frac{t_s}{2} \right) r_1}{n_s t_s L_s}. \quad (3.6)$$

In this study, because $r_1 = 20$ mm, $n_s = 9$, $t_s = 5.7$ mm and $L_s = 18.5$ mm, Sw is 1.84.

3.2.2 LES/NA-FGM

For numerical simulations of premixed combustion, premixed flamelet libraries based on the FGM [20], for which one-dimensional premixed free-propagating flame is calculated in several equivalence ratios, are often used. However, the conventional FGM has the deficiency that a set of chemical species' mass fractions at the equilibrium state of the reaction is basically stored in adiabatic states, so that the effect of heat loss on the change in the composition of chemical species cannot be considered. In this study, therefore, a non-adiabatic procedure, which was proposed by Kishimoto et al. [22] for generating non-premixed flamelet libraries that consider the effects of heat losses based on the flamelet/progress-variable approach [23], is employed for generating the premixed flamelet library. The calculations of the one-dimensional free-propagating flames for generating the premixed flamelet libraries for the FGM and NA-FGM are conducted by using FlameMaster [24], for which the non-adiabatic procedure [22] is introduced. The numbers of grids set for the mixture fraction, $\tilde{Z}$, product mass fraction, $\tilde{C}$ ($C = Y_{H_2} + Y_{H_2O} + Y_{CO} + Y_{CO_2}$), product mass fraction variance, $\tilde{C''}$, and enthalpy differences between with and without heat losses, $\Delta\tilde{h}$, are $100 \times 100 \times 50 \times 4$. Here, $\tilde{\phi}$ denotes Favre averaging of a physical quantity ϕ .

The governing equations for the LES employing the FGM and NA-FGM are the same. They are the conservation equations of mass, momentum, enthalpy $\tilde{h}$, mixture fraction $\tilde{Z}$, and progress variable $\tilde{C}$, as follows:

$$\frac{\partial \bar{\rho}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}}) = 0, \quad (3.7)$$

$$\frac{\partial \bar{\rho} \tilde{\mathbf{u}}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{\mathbf{u}}) = -\nabla \bar{P} + \nabla \cdot \bar{\boldsymbol{\sigma}} + \nabla \cdot \boldsymbol{\tau}, \quad (3.8)$$

$$\frac{\partial \bar{\rho} \tilde{h}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{h}) = \nabla \cdot (\bar{\rho} \tilde{D}_h \nabla \tilde{h}) + \nabla \cdot \mathbf{q}_h + Q_{\text{rad}}, \quad (3.9)$$

$$\frac{\partial \bar{\rho} \tilde{Z}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{Z}) = \nabla \cdot (\bar{\rho} \tilde{D}_Z \nabla \tilde{Z}) + \nabla \cdot \mathbf{q}_Z. \quad (3.10)$$

$$\frac{\partial \bar{\rho} \tilde{C}}{\partial t} + \nabla \cdot (\bar{\rho} \tilde{\mathbf{u}} \tilde{C}) = \nabla \cdot (\bar{\rho} \tilde{D}_C \nabla \tilde{C}) + \nabla \cdot \mathbf{q}_C + \bar{\rho} \tilde{\omega}_C, \quad (3.11)$$

Here, $\mathbf{u}$ is the velocity, P is the pressure, $\boldsymbol{\sigma}$ is the stress tensor, D_h , D_Z , and D_C are the diffusion coefficients of h , Z , and C , respectively, τ is the subgrid-scale stress term derived from the turbulence model, and $\mathbf{q}_h$, $\mathbf{q}_Z$, and $\mathbf{q}_C$ are the subgrid-scale terms for each scalar. Q_{rad} is the source term of radiative heat transfer. $\bar{\varphi}$ and $\tilde{\varphi}$ denote the LES filtering and Favre averaging of a physical quantity φ , respectively. The reaction rate of C , ω_C , and the mass fractions of chemical species i , Y_i , needed for calculating the temperature field are taken from the flamelet library, described above.

3.2.3 Computational details

The governing equations are solved using an unstructured LES solver: FrontFlow/Red extended by some research institutes, including Kyoto University, referred to as FFR-Comb [22, 25-27]. A dynamic Smagorinsky model [28] is used as the turbulence model. A second-order Euler implicit method is used for time advancement, and the time step is set to 1.0×10^{-5} s. The flamelet libraries for the FGM and NA-FGM are built using the mechanisms developed by the University of California San Diego [29] with nitrogen chemistry [30] in this study. This mechanism has 70 species and 321 reaction steps. The reason for selecting this mechanism is expressed in section 3.1. A gray gas model is adopted as the radiation model [31]. The absorption constant is 0.10, which is determined by reference to the results of RANS simulation with weighted sum of gray gasses model [32].

It is well-known that reactions related to NO are slow. NO concentration in the flamelet libraries is calculated at the equilibrium state. The reactions are often affected by heat losses in practical combustion chambers. In that case, the concentration is not coincident with experimental results. Due to solve this problem, Ihme and Pitsch's approach [33] is adopted. By preliminarily including the rates of production and consumption of NO in each flamelet library, NO concentration is calculated through the transport equation of NO in isolation with its concentration in the library. Therefore, for the LES/NA-FGM, the slow rate of the NO reactions in non-adiabatic states can be

considered. On the other hand, for the LES/FGM, the rate in an adiabatic state is considered.

3.3 Results and discussion

3.3.1 Validity of reaction mechanism

The predicted combustion behavior is strongly affected by the detailed reaction mechanism used in the LES. Therefore, in this section, the effects of the detailed reaction mechanism on the laminar burning velocity, S_L , and NO emissions are investigated in terms of one- and two-dimensional simulations, respectively. As the detailed reaction mechanism, GRI-mech 3.0, and the mechanisms developed by the University of California San Diego [29] with nitrogen chemistry [30], Tian et al. [34], and Okafor et al. [35] are featured. Here, these mechanisms are respectively referred to as GRI-mech, UCSD-mech, Tian-mech, and Okafor-mech which was developed by combining GRI-mech and Tian-mech.

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One-dimensional adiabatic and unstretched laminar premixed flames of ammonia/methane/air mixtures are calculated using ANSYS Chemkin-PRO [36]. S_L is defined as the velocity at the cold boundary in the computational domain.

Figure 3.2 shows the comparison of S_L obtained by one-dimensional calculations using different detailed reaction mechanisms, together with experiments of Okafor et al.[35]. The fuel consists of 52.1% NH_3 and 47.9% CH_4 by volume, and this constituent is close to that in the swirl burner. Tian-mech overestimates S_L in the whole ϕ range. Although GRI-mech shows good accuracy, the reactions related to NH_2 , which is an important species for the NO reactions, are not included [37]. Okafor-mech shows a good agreement in a wide range of ϕ . Finally, although UCSD-mech matches the experimental results at $\phi > 1$, it somewhat overestimates at $\phi < 1$.

Two-dimensional RANS simulations are conducted in order to compare the amount of NO emissions using each reaction mechanism. There are no previous studies that evaluated the amount of NO emissions for ammonia/methane/air laminar flames, so the experiment conditions by Valera-Medina et al. [14] are taken up in order to evaluate each

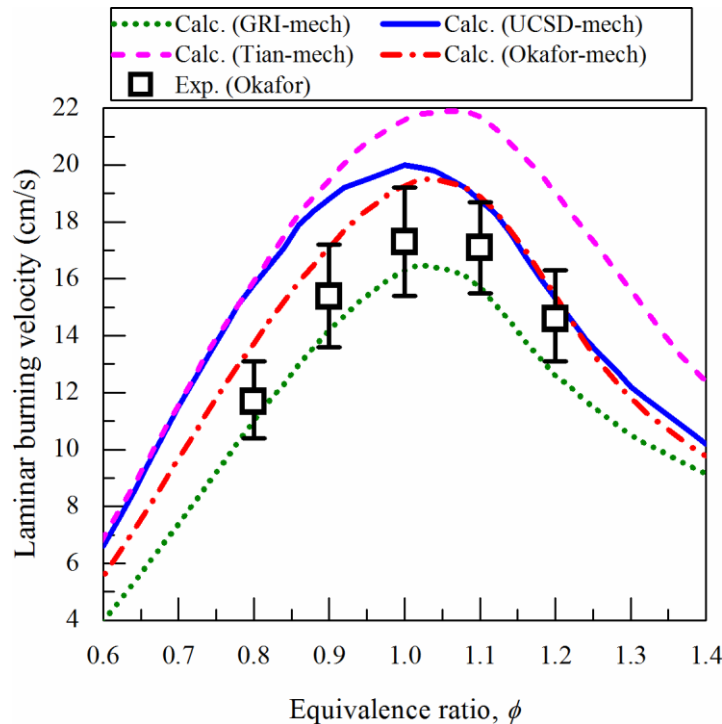


Figure 3.2 Comparison of laminar burning velocities obtained by one-dimensional calculations using different detailed reaction mechanisms, together with experiments.

reaction mechanism. Figure 3.3 shows the schematic of computational domain for two-dimensional RANS simulations. The domain is divided into ~ 90 thousand cells. As the conditions regarding the composition of the premixed gases, its flow rate and temperature and pressure are the same in section 3.1. For the premixed gas, the uniform radial velocity, V , and uniform circumferential velocity, W , are used as the inlet boundary condition. V and W are respectively expressed as

$$V = \frac{Q}{2\pi r_1 L_s}, \quad (3.12)$$

$$W = \frac{Q}{n_s t_s L_s}. \quad (3.13)$$

Wall conditions are adiabatic and any radiation model is not employed, due to focus on the elementary adequateness of each reaction mechanism for the swirl burner. Realizable k - ε model [39] is employed as a turbulence model.

Figure 3.4 shows the comparison of temperature distributions obtained by two-dimensional RANS simulations using different detailed reaction mechanisms at $\phi = 0.840$, 1.04 and 1.31, respectively. The distributions by GRI-mech, UCSD-mech and Okafor-mech are quite similar at each ϕ condition. There is great difference for only Tian-mech.

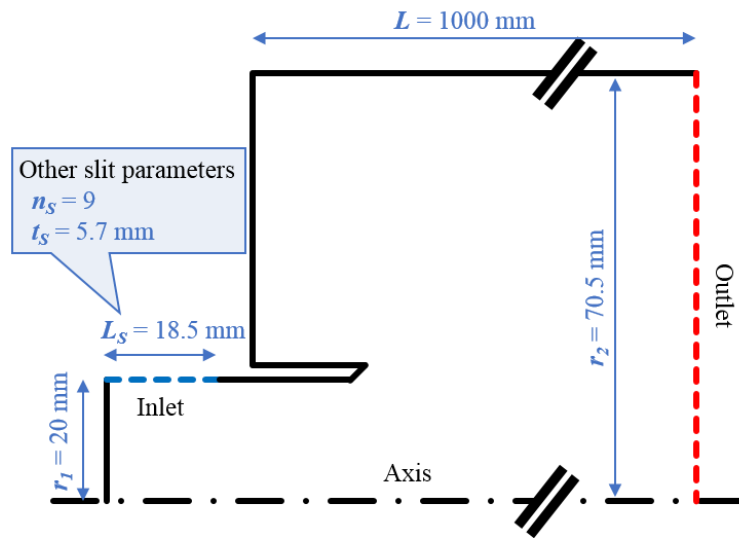


Figure 3.3 Schematic of computational domain for two-dimensional RANS simulations.

Although Tian-mech overestimates laminar burning velocity, the flame is lifted up from the bottom of the burner. In the experimental study, the actual flame was not lifted up at $\phi = 1.31$ [14]. Therefore, this shows Tian-mech is incompatible with the conditions of this study.

Figure 3.5 shows the comparison of the amounts of NO emissions obtained by two-dimensional RANS simulations using different detailed reaction mechanisms, together with experiments [14]. The NO concentrations for the whole conditions by the

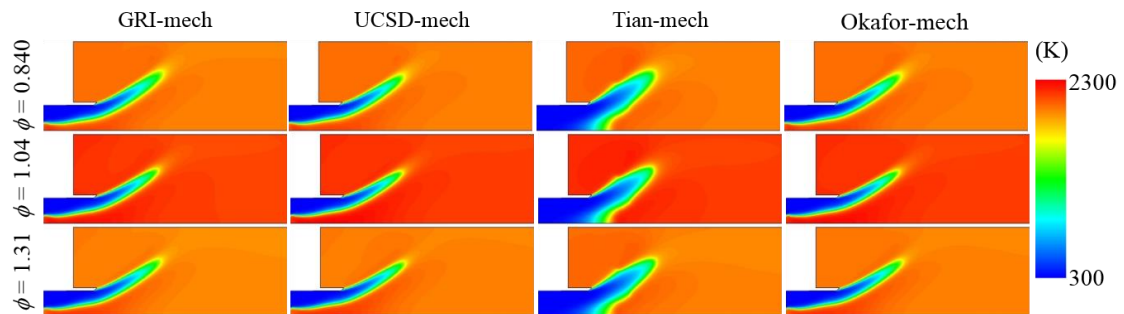


Figure 3.4 Comparison of instantaneous temperature distributions obtained by two-dimensional RANS simulations using different detailed reaction mechanisms at $\phi = 0.840, 1.04$ and 1.31 .

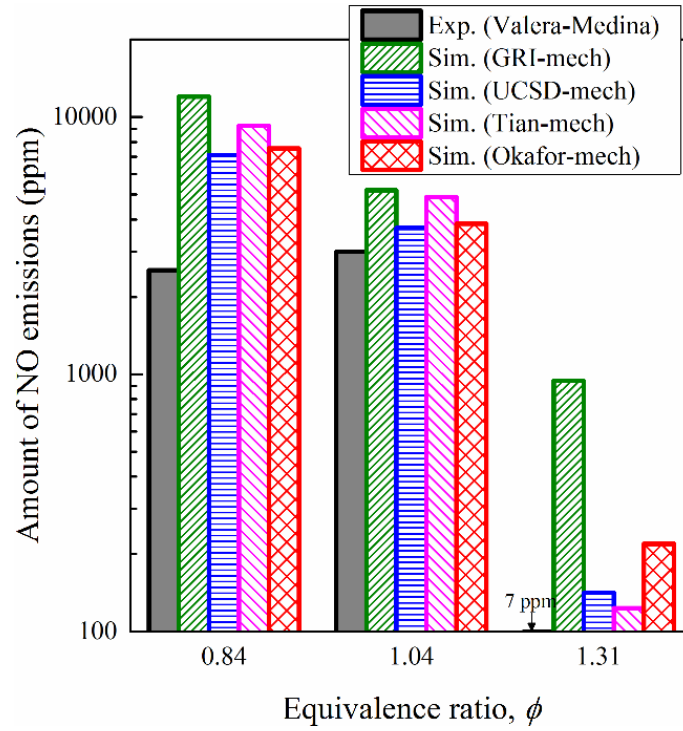


Figure 3.5 Comparison of time-averaged amounts of NO emissions obtained by two-dimensional RANS simulations using different detailed reaction mechanisms, together with experiments [15].

simulations reach the steady state before approaching the outlet boundary. All mechanisms overestimate the amounts of NO emissions in the whole ϕ range in comparison with the experimental results. The amount of NO emissions using UCSD-mech is comparatively close to the experimental result except for Tian-mech. NO emissions with both of Okafor-mech and UCSD-mech are similar at $\phi = 0.840$ and 1.04, but the amount of NO emissions at $\phi = 1.31$ with Okafor-mech is 1.55 times larger than that with UCSD-mech.

As above, because UCSD-mech has a relatively reasonable agreement among these mechanisms, it is employed for generating the flamelet libraries and discussing the NO and CO reactions in non-adiabatic states.

3.3.2 Predictions of NO and CO emissions

(a) Predictions of NO emissions

First, Fig. 3.6 shows the comparisons of distributions of (a) temperature, (b) axial

3.3 Results and discussion

velocity, (c) OH mole fraction, and (d) NO mass fraction at $\phi = 1.31$ between the LES/FGM and the LES/NA-FGM. The maximum temperature for the LES/FGM is 1974 K, the same as the adiabatic flame temperature, T_{ad} . However, the LES/NA-FGM shows 1950 K, and it is lower than the T_{ad} by 24 K by heat losses. There are no differences in terms of flow field between these approaches. The difference in OH mole fraction distributions, which is one of the most important species for the ammonia combustion, should be addressed. Once OH is produced in the LES/FGM, the concentration remains high throughout the combustion process. However, OH in the LES/NA-FGM is produced only in the flame region, and, after leaving this region, it disappears. This is the reason that the production of NO in the LES/NA-FGM is lower than that in the LES/FGM. The effects of temperature and the presence of H and OH radicals are discussed in section 3.3.1. Figure 3.7 shows the comparisons of axial profiles of NO concentration and temperature at $\phi = 1.31$ between the LES/FGM and the LES/NA-FGM. NO concentrations for both approaches rapidly decrease through passing each flame zone. After that, the NO concentration for the LES/FGM remains almost the same in the downstream. This is explained by Fig. 3.8, which shows the correlation between production rate of NO and progress variable, C , at $\phi = 0.840, 1.04$ and 1.31 for $\Delta h = 0$ (FGM). The production rate of NO in the downstream of a flame is negative at only $\phi =$

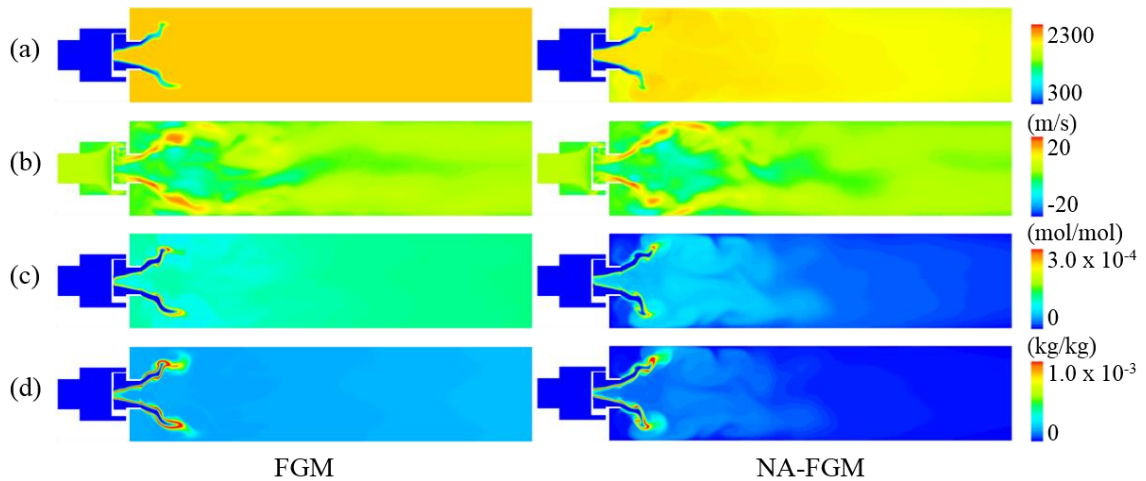


Figure 3.6 Comparisons of instantaneous distributions of (a) temperature, (b) axial velocity, (c) OH mole fraction, and (d) NO mass fraction at $\phi = 1.31$ between LES/FGM and LES/NA-FGM.

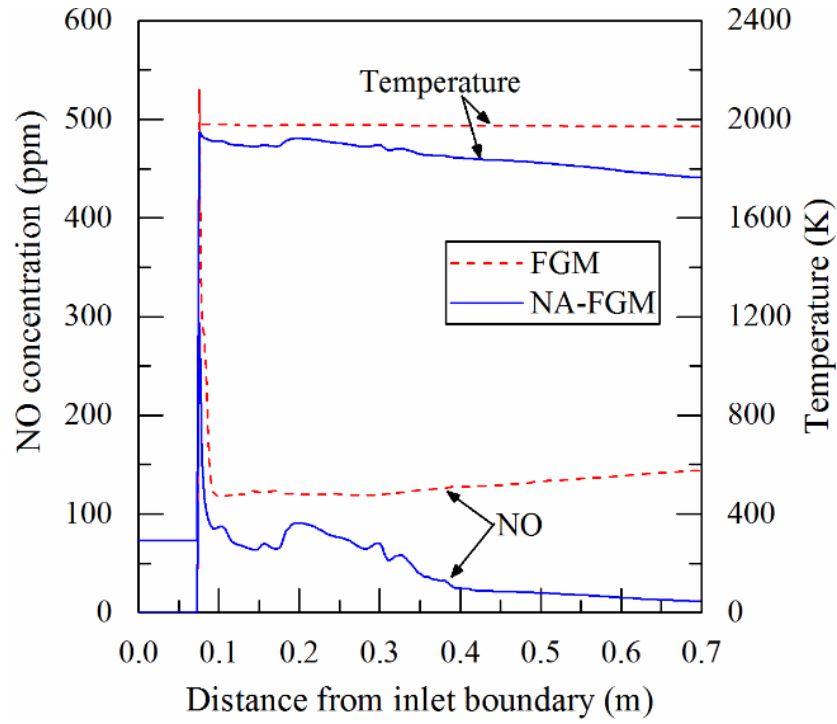


Figure 3.7 Comparisons of time-averaged axial profiles of NO concentration and temperature at $\phi = 1.31$ between LES/FGM and LES/NA-FGM.

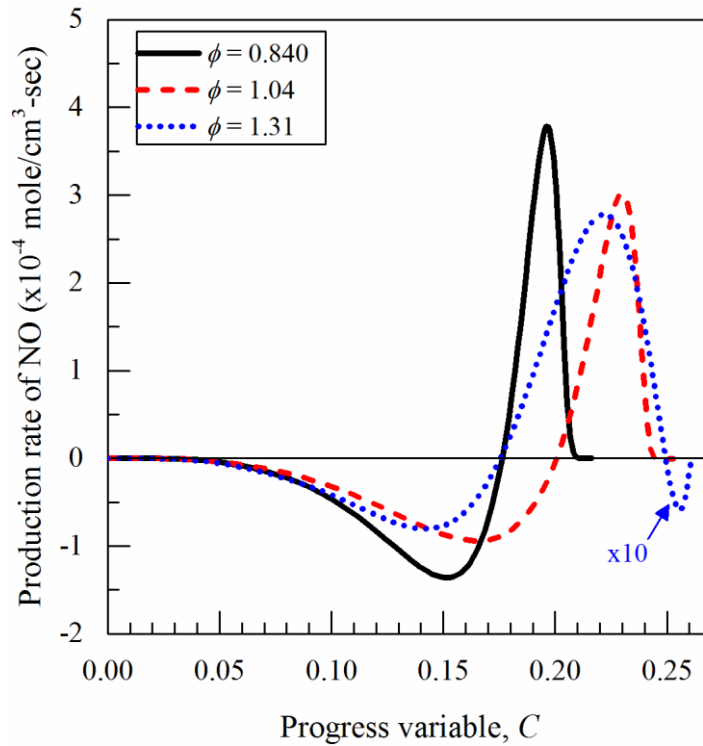


Figure 3.8 Correlation between production rate of NO and progress variable, C, at $\phi = 0.840, 1.04$ and 1.31 for $\Delta h = 0$ (FGM).

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1.31, and when the value of C becomes its maximum, the rate is almost zero. The LES/FGM estimates the amount of NO emissions as 148 ppm in Fig. 3.7. However, the NO concentration for the LES/NA-FGM monotonically decreases even in the downstream of a flame, and eventually reaches 9 ppm. This value is close to 7 ppm measured in the experiments. This shows the superiority of the LES/NA-FGM in estimating the amount of NO emissions, and it also shows the quantitative accuracy at $\phi = 1.31$.

Figure 3.9 shows the comparisons of distributions of (a) temperature, (b) axial velocity, (c) OH mole fraction, and (d) NO mass fraction for the LES/NA-FGM between the cases of $\phi = 0.840$ and 1.04. The maximum of OH mole fraction at $\phi = 0.840$ and 1.04 is larger by approximately 15 times than that at $\phi = 1.31$, increasing the reactivity at these ϕ conditions. As a result, the NO production is quite high, especially at $\phi = 0.840$. Each of NO concentrations at $\phi = 0.840$ and 1.04 for the LES/NA-FGM also decreases in the downstream of the flames. This is the peculiar behavior derived from the presence of heat losses, because the NO production rate in the downstream of a flame zones is almost zero in adiabatic states in Fig. 3.8.

Figure 3.10 shows the comparison of the amounts of NO emissions as a function of ϕ between the LES/FGM and the LES/NA-FGM, together with the results obtained by

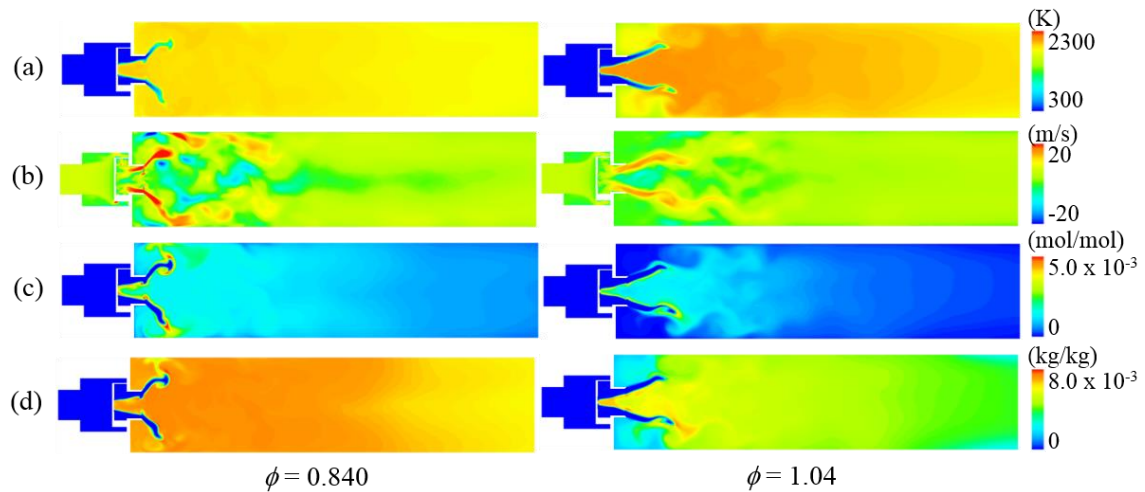


Figure 3.9 Comparisons of instantaneous distributions of (a) temperature, (b) axial velocity, (c) OH mole fraction, and (d) NO mass fraction for LES/NA-FGM between the cases of $\phi = 0.840$ and 1.04.

3.3 Results and discussion

one-dimensional premixed flame calculations (PREMIX) and experiments [15]. For comparison, the NO concentrations at the outlet boundary for one-dimensional laminar premixed flames are also illustrated as PREMIX. The amount of NO emissions for the LES/FGM is proximate to that for the PREMIX and is higher than that of the experiment for whole ϕ conditions, whereas that for the LES/NA-FGM is estimated quantitatively at $\phi > 1.04$. However, it is overestimated by more than twice, at $\phi = 0.840$, even when the LES/NA-FGM is employed. The reasons of the overestimation are as follows. First, the experiment at $\phi = 0.840$ may include an uncertainty. According to the experiment, the amount of O₂ emissions at $\phi = 0.840$ is approximately 4.5%. The concentration corresponds to that as $\phi = 0.752$ if the premixed gas is completely burned. However, because the amount of NO emissions at $\phi = 0.752$ for the LES/NA-FGM is still high in Fig. 3.10, it cannot be concluded that the experimental condition was equivalent to not as $\phi = 0.840$ but as $\phi = 0.752$. Second, some inaccuracies remain in UCSD-mech employed in this study. UCSD-mech overestimates S_L at $\phi < 1$, and the reactions concerning NO are

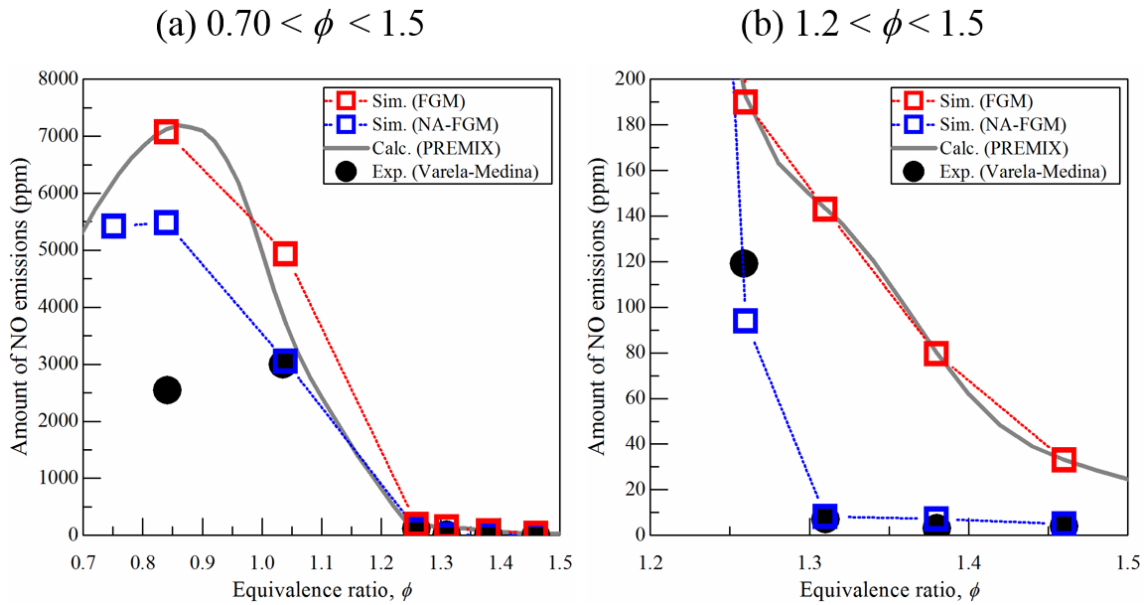


Figure 3.10 Comparison of time-averaged amounts of NO emissions as a function of ϕ between LES/FGM and LES/NA-FGM, together with the results obtained by adiabatic one-dimensional premixed flame calculations (PREMIX) and experiments. The display ranges are (a) $0.70 < \phi < 1.5$ and (b) $1.2 < \phi < 1.5$.

still not validated. At $\phi = 0.840$, there is very few effect on the consumption rate of NO in non-adiabatic states in section 3.3.1. If this should be the case, it means that UCSD-mech overestimates the NO production in a flame zone. For the quantitative prediction of NO emissions at $\phi = 0.840$, these unclear problems must be separately examined.

(a) Predictions of CO emissions

Because CO emissions are often problematic for practical uses because of its harmful effect on human bodies, this section deals with their evaluation. Figure 3.11 shows the comparisons of distributions of CO mole fraction at $\phi = 0.840$, 1.04 and 1.31 between the LES/FGM and the LES/NA-FGM. Once CO in the LES/FGM is produced at $\phi = 1.04$ and 1.31, the concentration remains high. This is the effect of thermal dissociation, and one part of CO_2 transforms into CO in high temperatures. This transformation is hardly seen in the LES/NA-FGM because of heat losses.

Finally, Fig. 3.12 shows the comparison of the amounts of CO emissions at $\phi = 0.840$ between the LES/FGM and the LES/NA-FGM, together with the experiments. The experimental results for other ϕ conditions were not accurately measured because of an upper limit of equipment range. At $\phi = 0.840$, the amount of CO emissions in the LES/FGM is overestimated, whereas that in the LES/NA-FGM is close to the experimental result, which emphasizes that the LES/NA-FGM is a more accurate tool to determine the amount of CO emissions.

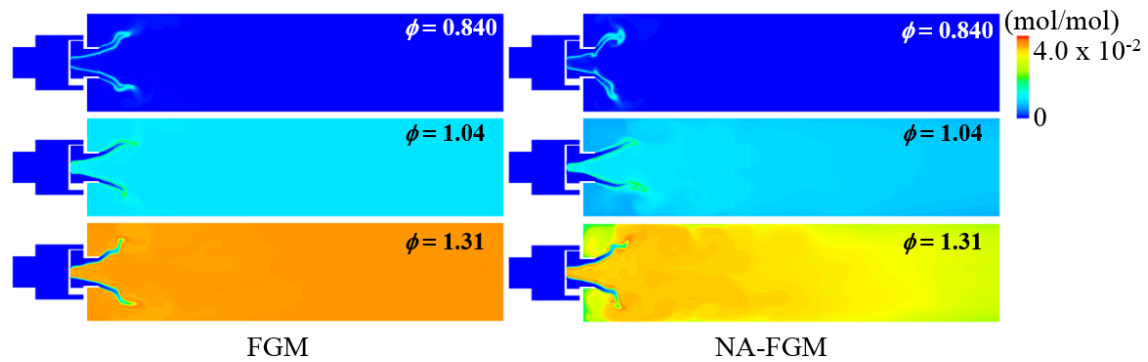


Figure 3.11 Comparisons of instantaneous distributions of CO mole fraction at $\phi = 0.840$, 1.04 and 1.31 between LES/FGM and LES/NA-FGM.

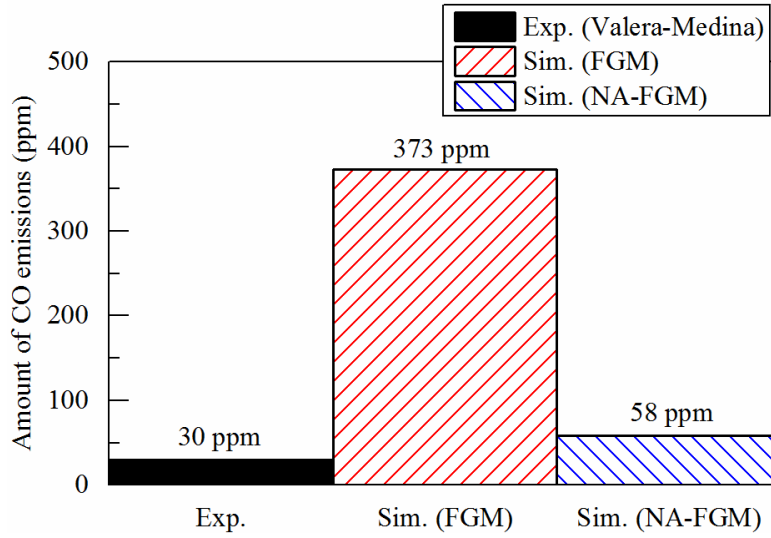


Figure 3.12 Comparison of time-averaged amounts of CO emissions at $\phi = 0.840$ between LES/FGM and LES/NA-FGM, together with experiments [15].

3.3.3 Effects of heat losses on NO and CO reactions

The effects of heat losses on the NO and CO reactions are investigated in detail in terms of one-dimensional premixed flame calculations below. The UCSD-mech is employed as the detailed reaction mechanism as ever. The fuel consists of 61.0% NH_3 and 39.0% CH_4 by volume, and the unburned temperature and pressure are 298 K and 1 atm, respectively.

(a) NO reactions

The reactions related to NO are slower than others. Therefore, it is assumed that heat losses affect the consumption rate of NO in the downstream region of flames. Figure 3.13 shows the time series of NO concentrations at $\phi = 0.840$, 1.04 and 1.31 for ammonia/methane/air combustion obtained by one-dimensional calculations in non-adiabatic states. Here, the downstream region of a flame is defined as the downstream side from the position that H mole fraction reaches the maximum. The all species' concentrations at the time in an adiabatic state are used as each initial condition. Then, the change of NO concentrations is calculated under constant temperature, T , in downstream regions. Besides, T_{ad} shows the adiabatic flame temperature at each ϕ

condition.

At $\phi = 0.840$, the NO concentrations are not changed other than the case of $T = 1900$ K, and it slightly decreases at $T = 1900$ K. At $\phi = 1.04$, the NO concentration at $T = T_{ad}$ remains over 4000 ppm. On the other hand, it dramatically decreases in non-adiabatic conditions, and especially at $T = 2000$ K, it eventually reaches 204 ppm. As temperature becomes lower than $T = 2000$ K, the consumption rate becomes slower, and the NO concentration at $T = 1500$ K rather increases and heads for 6300 ppm. That is, this value is larger than that in the adiabatic state. Finally, at $\phi = 1.31$, the NO concentrations become diminished as temperature decreases. In the case of $T = 1900$ K and 1800 K, the NO concentrations gradually decrease, and they eventually reach about 6 ppm. NO instantaneously disappears in the cases of $T < 1700$ K.

For comparison, Fig. 3.14 shows the time series of NO concentrations at $\phi = 0.840$, 1.04 and 1.31 for methane/air combustion obtained by one-dimensional calculations in non-adiabatic states. At $\phi = 0.840$ and 1.04, the production rate of NO increases through thermal NO_x processes as temperature becomes higher. For methane/air combustion, there is no specific temperature condition which strengthens the consumption of NO, and this trend is different from ammonia/methane/air combustion. On the other hand, the NO concentration decreases slowly at $\phi = 1.31$, and the consumption rate of NO becomes max at around 1900 K.

For ammonia/methane/air combustion, Fig. 3.15 shows the relevant reaction path related to the NO concentration at $\phi = 1.31$ at the time that its consumption rate is max by one-dimensional calculation in a non-adiabatic state. The ambient temperature is $T = 1900$ K. The thickness of each arrow denotes the relative magnitude of the reaction rate. NO is mainly consumed through three routes, or N₂O, N₂H and N₂ route respectively.



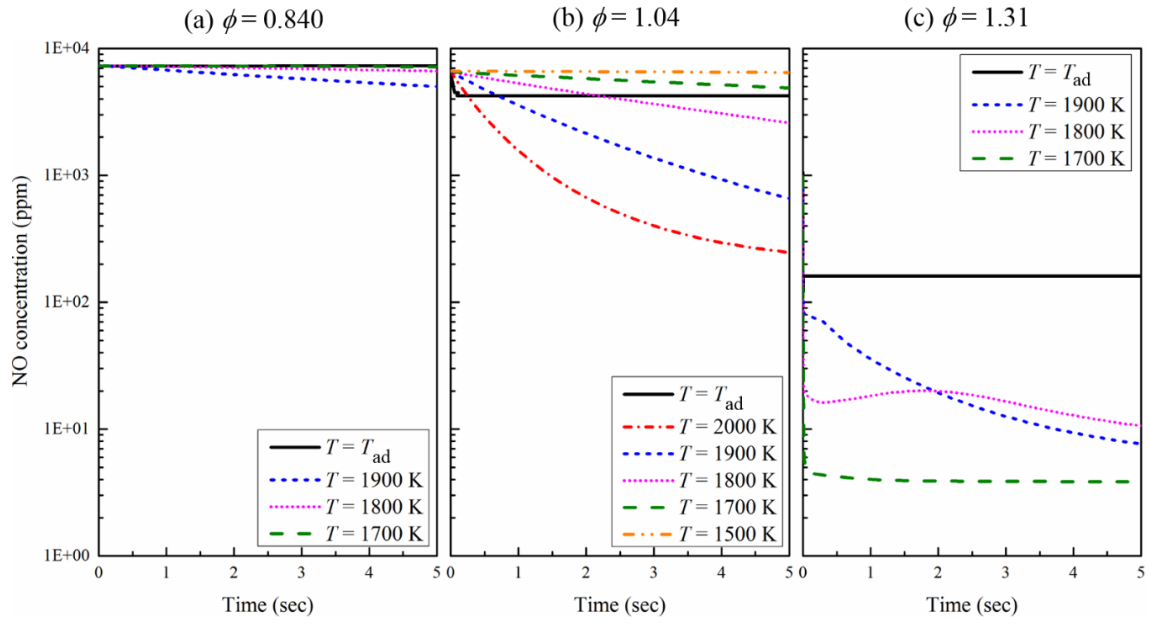


Figure 3.13 Time series of NO concentrations at $\phi = 0.840$, 1.04 and 1.31 for ammonia/methane/air combustion obtained by one-dimensional calculations in various temperature conditions.

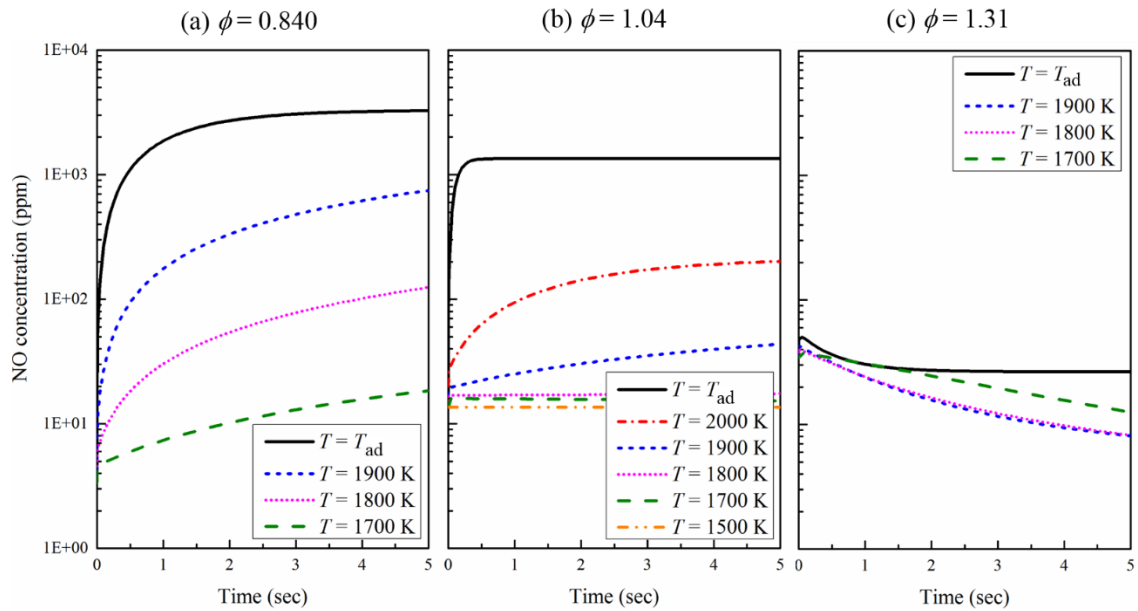


Figure 3.14 Time series of NO concentrations at $\phi = 0.840$, 1.04 and 1.31 for methane/air combustion obtained by one-dimensional calculations in various temperature conditions.

3.3 Results and discussion

In the N_2O route, N_2O changes to N_2 by reacting with H or OH radical. Moreover, NH_2 and NH , which are needed for above reactions, are generated from NH_3 and NH_2 , respectively. These reactions require H and OH radicals. Therefore, it is important for the consumption of NO that there are active radicals such as O and OH in non-adiabatic states.

As above, the effects of heat losses on the consumption of NO are not almost observed at $\phi = 0.840$, but the NO concentration slightly decreases at only $T = 1900$ K. At $\phi = 1.04$, NO concentration decreases the fastest at $T = 2000$ K, and as temperature becomes smaller, it rather increases. At $\phi = 1.31$, NO concentrations at $T = 1900$ K and 1800 K decrease gradually to about 6 ppm, and in the cases of $T < 1700$ K, it instantaneously disappears. Such heat losses and active radicals such as H and OH support

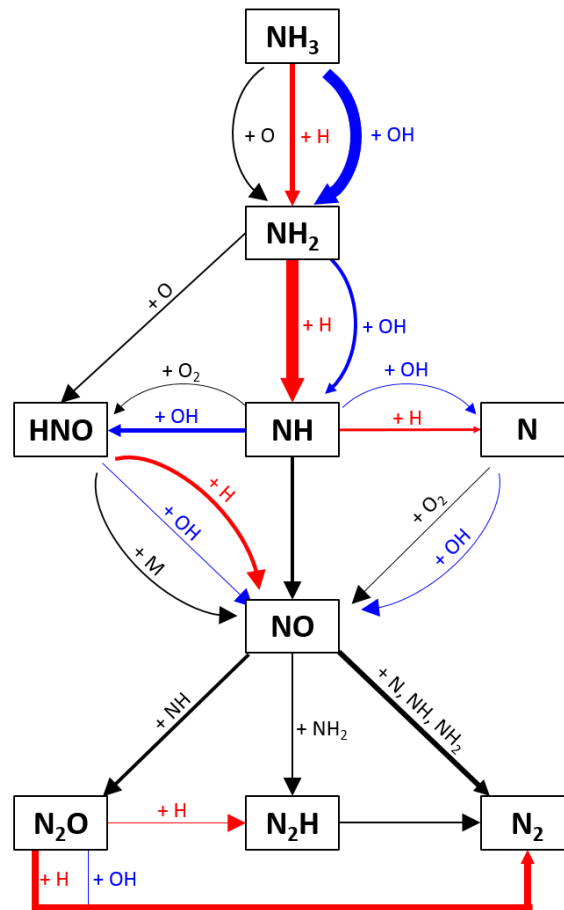


Figure 3.15 Relevant reaction path related to NO concentration at $\phi = 1.31$ for ammonia/methane/combustion at the time that its consumption rate is max by one-dimensional calculation in $T = 1900$ K.

the consumption of NO.

(a) CO reactions

Figure 3.16 shows the principal reactions of CO at $\phi = 0.840$ for ammonia/methane/air combustion obtained by one-dimensional calculation in an adiabatic state. The consumption rate of CO basically depends on one reaction,



OH radical is namely the most important species.

In order to evaluate the effects of heat losses, ambient temperature dependence for the consumption of CO is investigated. Figure 3.17 shows the time series of CO concentrations at $\phi = 0.840$ for (a) ammonia/methane/air combustion and (b) methane/air combustion, which are obtained by one-dimensional calculations in non-adiabatic states.

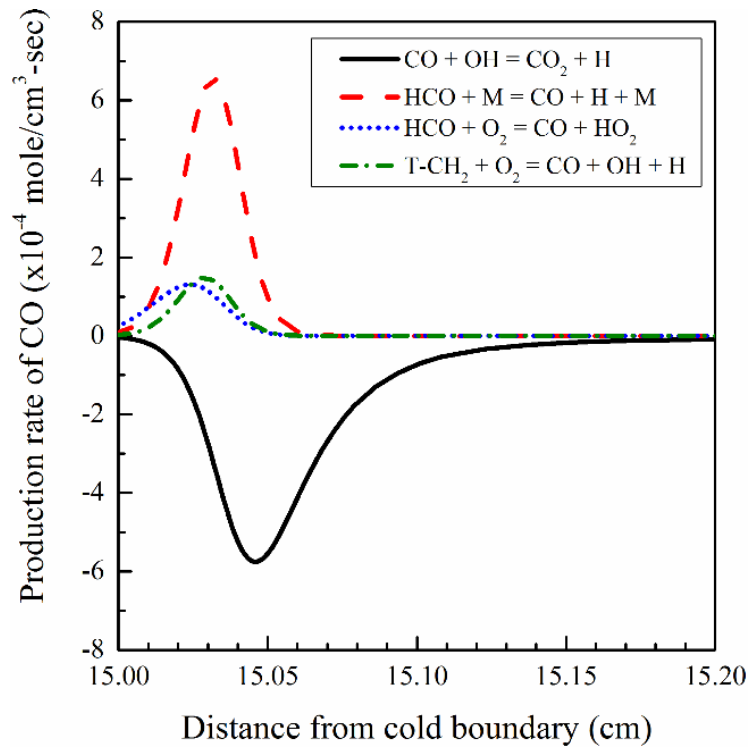


Figure 3.16 Principal reactions of CO at $\phi = 0.840$ for ammonia/methane/air obtained by one-dimensional calculation in an adiabatic state.

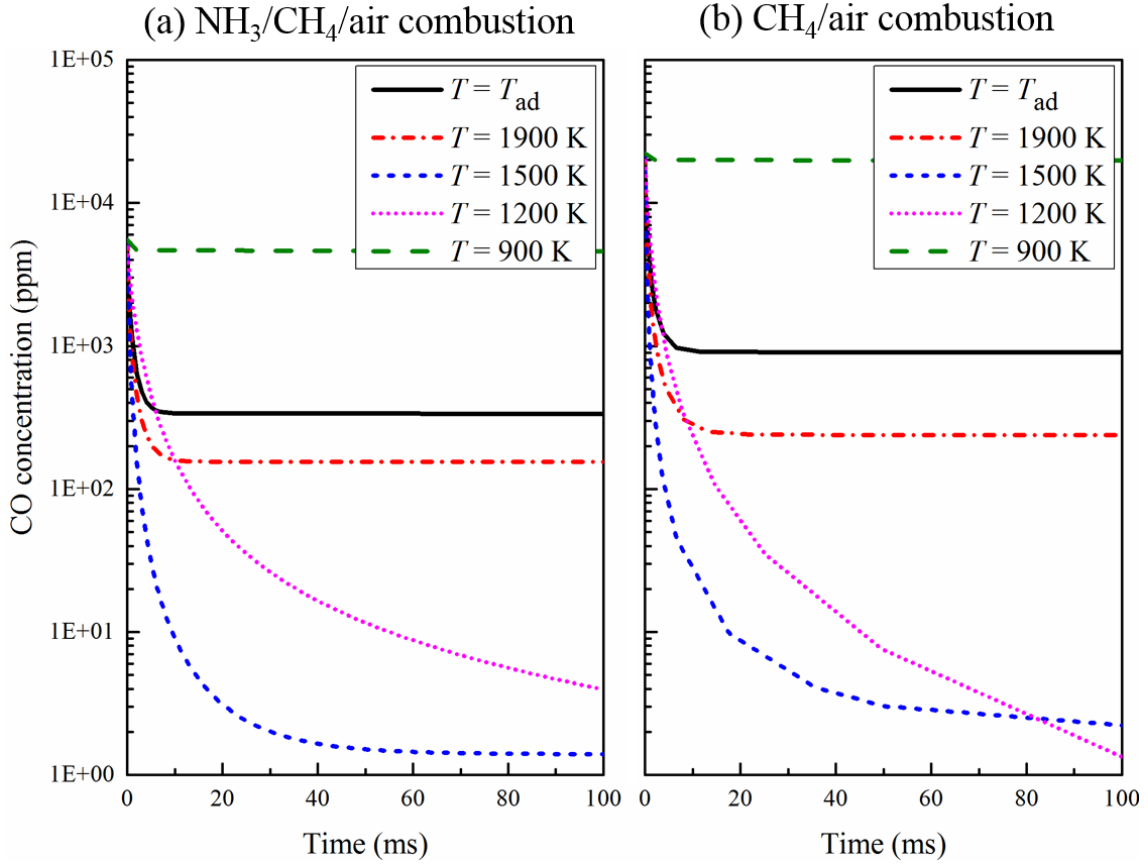


Figure 3.17 Time series of CO concentrations at $\phi = 0.840$ for (a) ammonia/methane/air combustion and (b) methane/air combustion obtained by one-dimensional calculations in various temperature conditions.

The definition of the downstream region is the same as the previous section. For ammonia/methane/air combustion, the consumption rate of CO becomes larger with rich OH radicals under low ambient temperature. As temperature becomes lower than T_{ad} , the consumption rate of CO becomes faster, and the rate becomes max around 1500 K. At $T = 1500$ K, the concentration reaches a few ppm within 40 ms. The consumption rate at $T = 1200$ K becomes slower than that at $T = 1500$ K, but the concentration reaches under 1 ppm after the passage of 200 ms. Because the required time is much larger than the reaction time of other main species, the transport equation of CO must be additionally employed in LES for the prediction of CO emissions in such non-adiabatic states. In other words, in this study, it is assumed that the amount of CO emissions can be evaluated without its transport equation. At $T = 900$ K, CO reactions are completely stopped and the concentration remains quite high. Additionally, the behavior of the CO concentration

for methane/air combustion is almost the same as that for ammonia/methane/air combustion.

3.4 Conclusions

In this chapter, the LESs/NA-FGM, which can account for the effects of various heat losses caused by radiation and cold walls, was applied to ammonia/methane premixed swirl combustion fields, and the formation mechanisms of NO and CO for ammonia combustion were investigated in detail. The NO and CO emissions for various equivalence ratios, ϕ , were compared with those predicted by the LESs/FGM and measured in the bespoke experiments [14]. The main results obtained in this chapter are summarized as follows.

- (1) The NO and CO emissions predicted by the LES/NA-FGM agree well with the experiments much better than the LES/FGM. This is because the NO and CO reactions for ammonia/methane/air combustion are quite susceptible to H and OH radicals' concentrations and gas temperature. This suggests that it is essential to take into account the effects of various heat losses caused by radiation and cold walls in predicting the NO and CO emissions for the combustion of ammonia as a primary fuel.
- (2) The NO emissions are successfully predicted using the LES/NA-FGM for $\phi > 1.04$, whereas it is overestimated at $\phi = 0.840$. This is considered to be due to the UCSD-mech used for generating the flamelet libraries. This mechanism tends to overestimate S_L , which may produce NO excessively in a flame zone.

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Chapter 4: Ammonia/city gas MILD combustion

4.1 Introduction

As mentioned in the previous chapter, ammonia (NH_3) is attracting attention not only as an energy carrier, but also as a carbon-free fuel that does not emit carbon dioxide (CO_2). Recently, many studies have been conducted on the direct combustion of fuels consisting of ammonia and either coal or natural gas [1-4]. Here, the application of ammonia combustion to the manufacturing sector, which is the second highest CO_2 emission sector [5], is focused on.

In heat treatment processes of the manufacturing sector, efforts have been made to reduce fuel consumption and CO_2 emissions. For example, the insulation performance of industrial furnaces has been improved, and heat loss in exhaust gas has been reduced by introducing recuperators or regeneration systems. In addition, CO_2 emissions of approximately 30% are reduced by the change of fuel sources from oil to city gas. The city gas is one of important fuels in Japan. In particular, 13A is widely used as the most popular standard for the city gas, and its higher heating value (HHV) is adjusted to 45 MJ/m^3 as standard state by mixing hydrocarbons having many carbon atoms such as propane or butane with natural gas. As further efforts to reduce CO_2 emissions, ammonia combustion has been a focus of attention. Ammonia liquefies at only 8.5 bar and can be easily supplied to remote locations from liquefied natural gas bases, using regular lorries [6], making it much easier to handle than hydrogen. Also, it is easy to detect ammonia leaks because it releases an acrid odor, even when odorant is not added. As the disadvantages of ammonia compared to methane, it has toxicity and requires special handling, and a slower burning velocity, by one fifth, and exhibits low combustibility [7]. In addition, ammonia combustion tends to increase NO_x emissions, due to the presence of nitrogen in its molecule. Recently, Kobayashi et al. [4] studied extensively about ammonia combustion and showed low burning rates and low extinction limits of ammonia flames. In order to quantitatively evaluate NO_x emissions, new detailed and reduced chemical reaction mechanisms are also being actively developed [8, 9]. Above all, main

issues for ammonia combustion are the low combustibility and the reduction of NO_x emissions.

In this chapter, Moderate or Intense Low-oxygen Dilution (MILD) combustion technique [10], which is widely used in the manufacturing sector to reduce NO emissions, is also focused on. The MILD combustion can be achieved through fuel direct injection (FDI), in which fuel and oxidant are separately injected into a furnace. Large circulation regions are generated in the furnace, by blowing them at high speeds, and the burned gas circulates to dilute unburned fuel and oxidant, in a process known as exhaust gas recirculation (EGR), with the result that the diluted fuel and oxidant react slowly. The MILD combustion is sometimes called flameless combustion [11], as there is no difference of brightness between the flames and the furnace walls, making it impossible to check flames visually. In this study, MILD combustion, flameless combustion, or high temperature air combustion [12] is treated as synonymous. In the MILD combustion, rapid temperature rise is suppressed because the heat generation rate approaches the heat supply rate for the object being heated. Ben-Mansour et al. [13] researched oxyfuel combustion and air–fuel combustion in a typical furnace. They indicated that the furnace temperature levels were greatly reduced as the percentage of recirculated CO₂ is increased. As the results, the production of thermal NO_x is suppressed. In addition, the MILD combustion is compatible with waste heat recovery technology, such as regeneration systems that preheat air using high-temperature burned gas. The thermal properties of the regenerative system are complex and one of research subjects. Wołkowycki [14] provided detailed experimental data with an industrial-scale glass-melting furnace. When a regeneration system is introduced, flame temperatures increase by an amount correlated to the amount of preheated energy, and a large amount of NO are usually produced. However, both saving energy consumption and reducing NO emissions can be achieved by combining the MILD combustion techniques by FDI with regeneration systems. The MILD combustion has been proven in real factories and has been studied in even recent years. There are lots of kinetic studies on methane as fuel in particular [15-17]. As other examples, the MILD combustion of a premixed methane jet in hot co-flow was investigated through comparing a typical bluff-body flame [18]. For the MILD combustion, laminar flame speed was very small and far below the local flow speed. And chemical and mixing times were compatible so that the Damkohler number was close to

1.0. Mardani et al. [19] evaluated the changes in MILD conditions due to the addition of hydrogen to methane and syngas. Syngas showed more sensitivity to hydrogen enrichment than methane, which made hydrogen a good additive to syngas in terms of MILD condition benefits. However, there have been no studies on whether MILD combustion contributes to the reduction of NO emissions for ammonia/city gas combustion.

The purpose of this study is therefore to investigate the effectiveness of MILD combustion in reducing NO emissions for ammonia and the city gas co-combustion. The fuels consist of ammonia and city gas, and the NH_3 concentrations in the fuels at lower heating value basis (LHV) are changed as a parameter. Zero-dimensional and three-dimensional numerical simulations are conducted, using detailed chemical reaction mechanisms. In the zero-dimensional simulations, the effects of the NH_3 concentration in the fuel at LHV basis, E_{NH_3} , the excess air ratio, λ , temperature, T , and EGR dilution rate, δ , on NO emissions are investigated. In this work, E_{NH_3} is defined as follows:

$$E_{\text{NH}_3} = \frac{LHV_{\text{NH}_3} Q_{\text{NH}_3}}{LHV_{\text{NH}_3} Q_{\text{NH}_3} + LHV_{13\text{A}} Q_{13\text{A}}}, \quad (4.1)$$

where LHV_φ and Q_φ are the LHV and volume flow rate of φ , respectively. λ represents the excess air ratio and is the reciprocal of the equivalence ratio. δ is defined as the rate at which the burned gas diluted the fuel and air. That is, $\delta = 0\%$ and $\delta = 100\%$ indicate no dilution and burned gas only, respectively. In the three-dimensional numerical simulations of an actual furnace employing the FDI and a regeneration system, the validity of the three-dimensional numerical simulation is examined by comparing the simulation results with the experimental results for city gas combustion. Then, for $E_{\text{NH}_3} = 20\%$ and 100% , the effects on NO emission reduction of MILD combustion, and burner design parameters for reducing NO emissions are investigated.

4.2 Numerical and experimental methods

4.2.1 Targeted experiments

Figure 4.1 shows the targeted furnace, which has inner dimensions of $3,500 \times 1,200$

$\times 1,200$ mm (Length $\times$ Width $\times$ Height). The furnace outer periphery is surrounded by

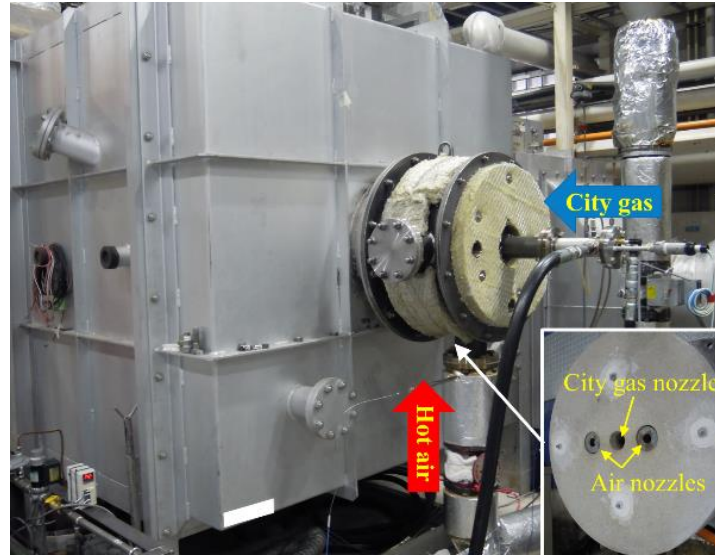


Figure 4.1 Targeted furnace and burner unit.

heat-resistant bricks, and it normally operated at under $1,300^{\circ}\text{C}$. The furnace has a $\phi 500$ mm opening in its front face, and a burner unit supplying fuel and air is mounted there. The burner unit is surrounded by castable refractories and has seven holes inside. A pilot burner is a double tube burner and is installed in the center hole of the burner unit, and the fuel and air are supplied through the inner tube and outer tube, respectively. The each volume flow rate is $10 \text{ m}^3/\text{h}$ at standard state, indicating $\lambda \sim 0.09$ and the fuel rich condition. λ means the ratio of actual air-fuel-ratio to stoichiometry for the fuel and air mixture. In normal operations, the fuel and air mixture is reacted, and the flame is held in a pilot burner's cup. The remaining fuel-rich mixture is blown into the furnace. Here, the pilot burner nozzle is referred to as simply a fuel nozzle. Two air nozzles are provided symmetrically across the fuel nozzle, and the distances between the air nozzles and the fuel nozzle, L , are 75, 130 or 190 mm. The other four nozzles are unused and filled with castable refractories. The inner diameter of the air nozzles can be adjusted by inserting metal cylinders, with diameter options of 19.4, 25.0, 35.5 or 50.0 mm. After the fuel and air mildly combust, the burned gas is discharged from a flue provided on the opposite side of the front face.

The fuel used is city gas 13A. At the time of our study, ammonia could not be supplied to the furnace, as there was no supply equipment.

The fuel and air flow rates are controlled by each mass flow meter, with the both

flow rates of the fuel and air supplied to the pilot burner set to $10 \text{ m}^3/\text{h}$, respectively. The remaining air is heated to 500°C , and is supplied from the two air nozzles. In order to simulate the introduction of the regeneration system, a separate air pre-heating furnace is used, allowing air supply at a temperature controlled to 500°C . Here, λ means the excess air ratio of the total air supply amount with respect to the fuel supply amount. Figure 4.2 shows the positions of the thermocouples installed for measuring furnace temperature. We use R-type thermocouples with each measurement performed at a point 100 mm below the furnace ceiling, with the exception of T11, which is measured at a point 600 mm below the ceiling. The burned gas components are measured at the flue using a controlled potential electrolysis sensor or a galvanic cell sensor. Measurement species are nitric oxide, nitrogen dioxide, carbon monoxide, carbon dioxide, unburned hydrocarbon and oxygen.

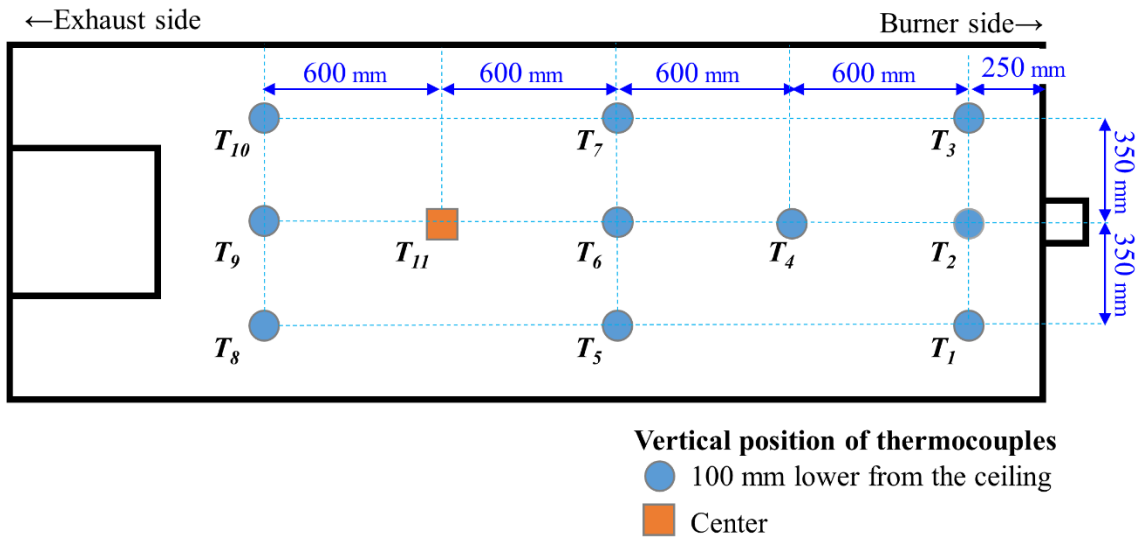


Figure 4.2 Positions of thermocouples installed for measuring furnace temperature.

4.2.2 Zero-dimensional kinetic simulations

ANSYS CHEMKIN PRO 17.2 [20] is used to conduct our zero-dimensional kinetic simulations. A closed homogeneous reactor model is used. Assuming MILD combustion, NO production is investigated in changing NH_3 concentration in the fuel, E_{NH_3} , excess air ratio, λ , temperature, T , and dilution rate by EGR, δ , at constant temperature and pressure.

The fuel used is a mixture of ammonia and city gas. Here the city gas composition is defined as 83.5% CH₄ and 16.5% C₂H₆ by volume. This ratio is set so that the HHV of the city gas is 45 MJ/m³ at standard state.

To define the concentrations of each species required for the simulation, if 13A is 1 mole, the molar quantities of reactant species i , n_i^r , are set as shown in Eqs. 4.2–4.6:

$$n_{\text{NH}_3}^r = 2.38 \frac{E_{\text{NH}_3}}{1 - E_{\text{NH}_3}}, \quad (4.2)$$

$$n_{\text{CH}_4}^r = 0.835, \quad (4.3)$$

$$n_{\text{C}_2\text{H}_6}^r = 0.165, \quad (4.4)$$

$$n_{\text{O}_2}^r = (0.75 n_{\text{NH}_3}^r + 2 n_{\text{CH}_4}^r + 3.5 n_{\text{C}_2\text{H}_6}^r) \lambda, \quad (4.5)$$

$$n_{\text{N}_2}^r = 3.76 n_{\text{O}_2}^r. \quad (4.6)$$

Here, these equations are invalid for $E_{\text{NH}_3} = 1$. In addition, chemical species, i , of the burned gas, n_i^b , is simply set as shown in Eqs. 4.7–4.10:

$$n_{\text{CO}_2}^b = 1.165, \quad (4.7)$$

$$n_{\text{H}_2\text{O}}^b = 1.5 n_{\text{NH}_3}^r + 2.165, \quad (4.8)$$

$$n_{\text{O}_2}^b = (\lambda - 1) n_{\text{O}_2}^r, \quad (4.9)$$

$$n_{\text{N}_2}^b = n_{\text{N}_2}^r + 0.5 n_{\text{NH}_3}^r, \quad (4.10)$$

where the value of λ is greater than 1. By introducing δ , to assume an EGR environment, initial mole fractions for each species i , X_i , are set as shown in Eqs. 4.11–4.17:

$$X_{\text{NH}_3} = (1 - \delta) \frac{n_{\text{NH}_3}^r}{\sum_i n_i^r}, \quad (4.11)$$

$$X_{\text{CH}_4} = (1 - \delta) \frac{n_{\text{CH}_4}^r}{\sum_i n_i^r}, \quad (4.12)$$

$$X_{\text{C}_2\text{H}_6} = (1 - \delta) \frac{n_{\text{C}_2\text{H}_6}^r}{\sum_i n_i^r}, \quad (4.13)$$

$$X_{O_2} = (1 - \delta) \frac{n_{O_2}^r}{\sum_i n_i^r} + \delta \frac{n_{O_2}^b}{\sum_i n_i^b}, \quad (4.14)$$

$$X_{N_2} = (1 - \delta) \frac{n_{N_2}^r}{\sum_i n_i^r} + \delta \frac{n_{N_2}^b}{\sum_i n_i^b}, \quad (4.15)$$

$$X_{CO_2} = \delta \frac{n_{CO_2}^b}{\sum_i n_i^b}, \quad (4.16)$$

$$X_{H_2O} = \delta \frac{n_{H_2O}^b}{\sum_i n_i^b}. \quad (4.17)$$

As detailed reaction mechanisms, those published by Okafor et al. [21], Tian et al. [22], the University of California at San Diego including nitrogen chemistry [23, 24], Mathieu and Petersen [25], and Nakamura and Hasegawa [26] are used for comparison. Okafor et al. [21] established the mechanism by combining GRI-mech 3.0 [27] with that of Tian et al. [22]. The mechanism of Nakamura and Hasegawa [26] does not include reactions related to hydrocarbons, so those of GRI-mech 3.0 excluding nitrogen chemistry is added. These mechanisms are referred to as Okafor-mec, Tian-mec, UCSD-mec, Mathieu-mec and Nakamura+GRI-mec, respectively.

4.2.3 Three-dimensional RANS simulations

Three dimensional RANS simulations are conducted using ANSYS FLUENT 17.2 [28]. This application has a long history in MILD combustion research, and Ishii et al. [29] studied the turbulent non-premixed combustion process for a regenerative gas-fired furnace, and compared the numerical simulation results and the experimental data. Figure 4.3 shows the computational domain which faithfully reproduced the actual furnace in Fig. 4.1, although our simulations are actually completed using a half model, in consideration of left–right symmetry. The burning capacity is 113 kW at LHV basis, and the fuel with E_{NH_3} and pilot air are supplied from the fuel and pilot air inlets, respectively. The city gas composition is as described in the Zero-dimensional RANS simulation, and the fuel and air temperatures and standard flow rates are 25°C and 10 m³/h. The test furnace has two main air inlets, from which the remaining air is supplied at 500°C. Total excess air ratio, λ , is set to 1.10. Our experimental results indicate that the average furnace

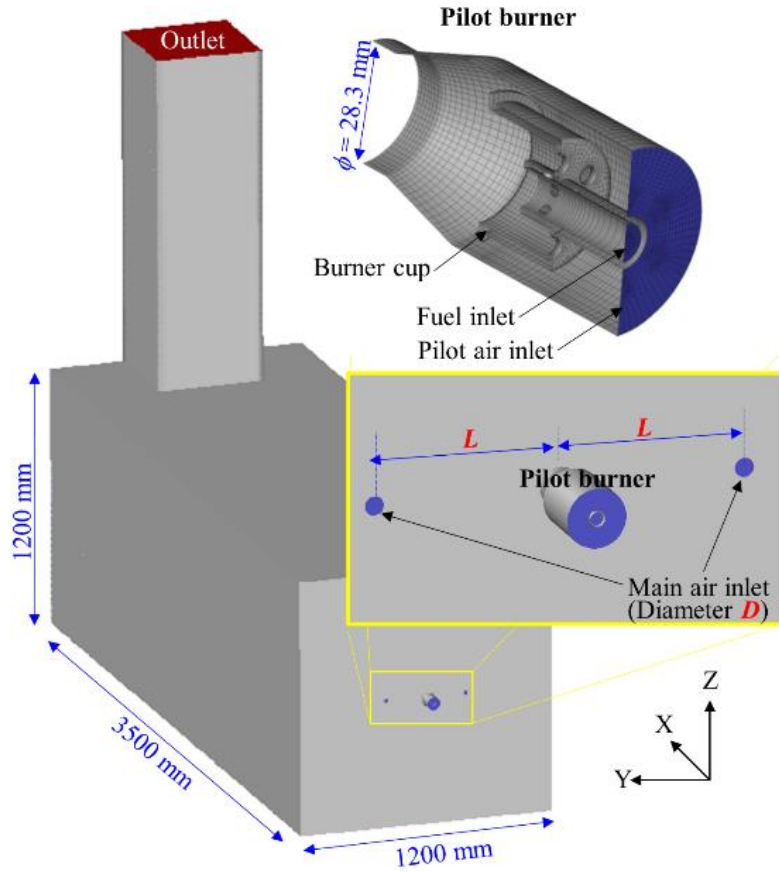


Figure 4.3 Computational domain for three-dimensional RANS simulations.

temperature is 1,250°C, and this is modulated by controlling the heat transfer coefficient on the furnace walls.

The Realizable $k-\varepsilon$ model [30] is used as our turbulence model, and the eddy dissipation concept model [31] with Okafor-mec is employed as our turbulence combustion model. As a radiation model, a discrete ordinates model applying the weighted sum of gray gas model [32, 33] is used.

In this study, multiple tests involving variations to E_{NH_3} , D , and L , are conducted. The E_{NH_3} conditions are 0%, 20% or 100%. Initially though, for the city gas combustion cases described in the Validity of RANS simulation, the simulation method validity is confirmed by comparing simulation results with experimental results. After that, the cases for ammonia and city gas co-combustion, and pure-ammonia combustion are also calculated. Table 1 shows typical parameter conditions, and a detailed review of results presented in the NO emissions for MILD combustion of ammonia / city gas.

4.3 Results and discussion

4.3.1 Sensitivity analysis on NO emissions

In this section, the results of the zero-dimensional kinetic simulations are focused on. Figure 4.4 shows NO concentrations after a residence time of 1 s against E_{NH_3} , calculated using various reaction mechanisms. The other parameters are fixed as $\delta = 0\%$, $\lambda = 1.10$, $T = 1250^\circ\text{C}$ and atmospheric pressure. For all the reaction mechanisms, NO concentrations tend to be upwardly convex with respect to E_{NH_3} , reaching maximums at $E_{\text{NH}_3} = 20\text{--}40\%$. For $E_{\text{NH}_3} = 20\%$, the NO concentrations, derived using Tian-mec and Nakamura+GRI-mec, exceed 10000 ppm, although when derived with the other mechanisms, the values are 6000–8000 ppm. For $E_{\text{NH}_3} > 40\%$, the NO concentrations dropped rapidly, and for $E_{\text{NH}_3} = 100\%$, the NO concentrations for most of the reaction mechanisms are less than 1100 ppm. The concentration for $E_{\text{NH}_3} = 100\%$, with only Nakamura+GRI-mec, is approximately 4500 ppm. Figure 4.5 shows NO concentrations after a residence time of 1 s against δ , calculated using various reaction mechanisms. The other parameters are fixed as $E_{\text{NH}_3} = 20\%$, $\lambda = 1.10$, $T = 1250^\circ\text{C}$ and atmospheric pressure.

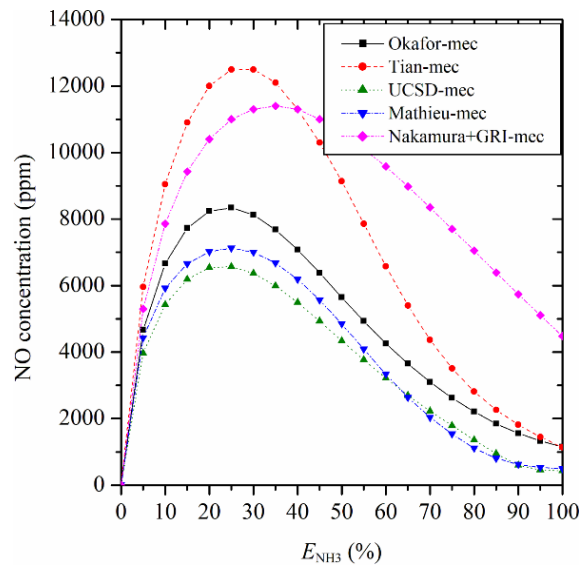


Figure 4.4 NO concentrations after a residence time of 1 s against E_{NH_3} , calculated using various reaction mechanisms. The other parameters are fixed as $\delta = 0\%$, $\lambda = 1.10$, $T = 1250^\circ\text{C}$ and atmospheric pressure.

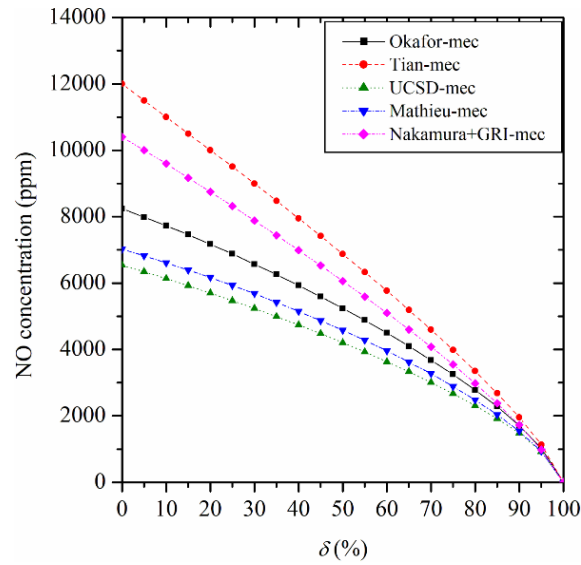


Figure 4.5 NO concentrations after a residence time of 1 s against δ , calculated using various reaction mechanisms. The other parameters are fixed as $E_{\text{NH}_3} = 20\%$, $\lambda = 1.10$, $T = 1250^\circ\text{C}$ and atmospheric pressure.

The NO concentrations significantly decrease as δ increases, for all the reaction mechanisms, which suggest that NO emissions fall less than 2000 ppm if $\delta > 90\%$ is achieved.

It is confirmed that NO concentration behavior, in response to E_{NH_3} and δ variation, is similar among all the reaction mechanisms. Therefore, Okafor-mec is used for the simulations involved in the following tests. Figure 4.6 shows NO emission amounts after a residence time of 1 s against temperature, for various E_{NH_3} , calculated using Okafor-mec. The other parameters are fixed as $\delta = 0\%$, $\lambda = 1.10$ and atmospheric pressure. The emissions are corrected at 11% O_2 , which is a correcting condition for evaluating NO_x emissions from industrial furnaces. The NO emissions increase with temperature. For $E_{\text{NH}_3} = 0\%$, the emissions are small in all areas. For $T < 1100^\circ\text{C}$, the emissions for $E_{\text{NH}_3} = 10\%$ are higher than those for the other concentrations, while for $E_{\text{NH}_3} = 20\text{--}40\%$ much higher values are recorded as the temperature increases. The emissions are relatively small for $E_{\text{NH}_3} = 100\%$, despite the large amount of N in the fuel.

Figure 4.7 shows NO emission amounts after a residence time of 1 s against λ , for various E_{NH_3} , calculated using Okafor-mec. The other parameters are fixed as $\delta = 0\%$, T

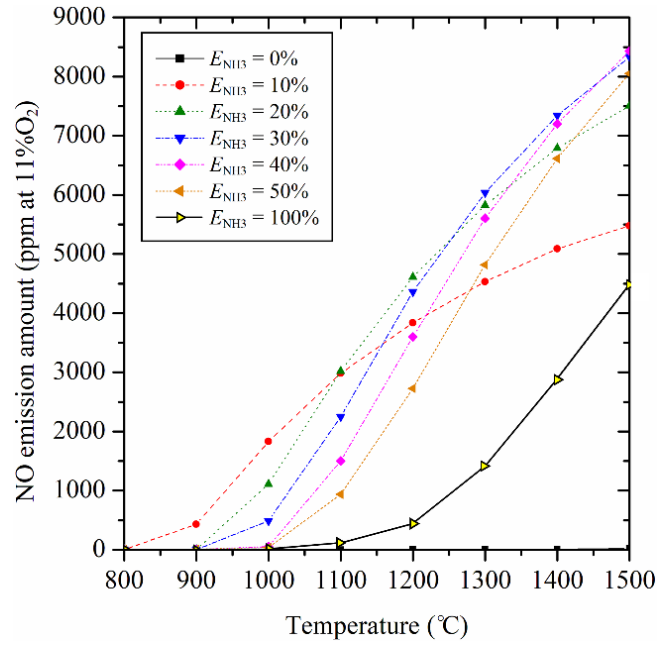


Figure 4.6 NO emission amounts after a residence time of 1 s against temperature, for various E_{NH_3} , calculated using Okafor-mec. The other parameters are fixed as $\delta = 0\%$, $\lambda = 1.10$ and atmospheric pressure. The emissions are corrected at 11% O₂.

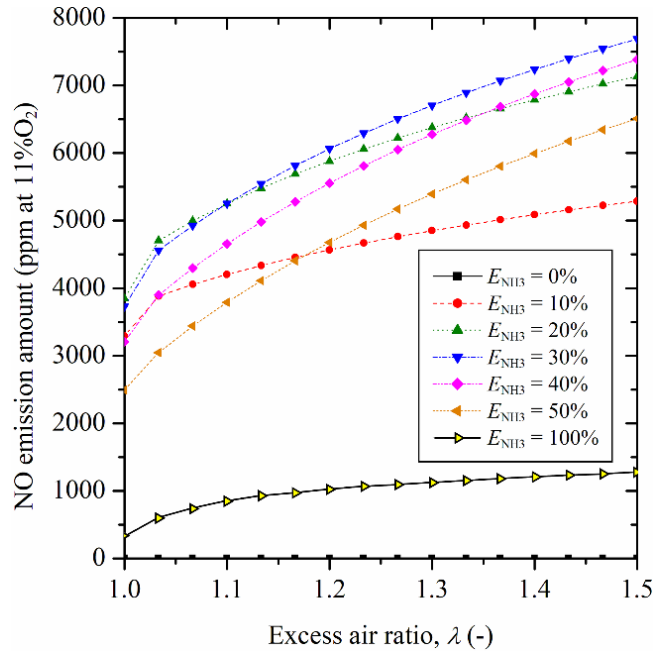


Figure 4.7 NO emission amounts after a residence time of 1 s against λ , for various E_{NH_3} , calculated using Okafor-mec. The other parameters are fixed as $\delta = 0\%$, $T = 1250^\circ\text{C}$ and atmospheric pressure. The emissions are corrected at 11% O₂.

= 1250°C and atmospheric pressure. The NO emissions increase monotonously with λ . When $E_{\text{NH}_3} = 20\text{--}50\%$, the slopes of the NO emission amounts with respect to λ are similar, while the slopes for $E_{\text{NH}_3} = 10\%$, and $E_{\text{NH}_3} = 100\%$ are quite gradual. Figure 4.8 shows the unburned fuel emission amounts after a residence time of 1 s against λ for various E_{NH_3} . Here, NH_3 , CH_4 , C_2H_6 and CO are defined as unburned components. The other conditions are the same as Fig. 4.7. The unburned fuel emission amounts are the highest around $E_{\text{NH}_3} = 20\%$. At $\lambda < 1.05$, the amounts increase drastically with decreasing λ . In order to make the amount less than 10 ppm, λ should be set to more than 1.05. These results indicate that, in order to reduce NO emissions without discharging unburned fuel, the appropriate λ range is 1.05–1.10.

Finally, the effects of EGR are subjected to additional investigation. Figure 4.9 shows NO emission amounts after a residence time of 1 s against δ , for various E_{NH_3} , calculated using Okafor-mec. The other parameters are fixed as $\lambda = 1.10$, $T = 1250^\circ\text{C}$ and atmospheric pressure. As with the results shown in Fig. 4.5, NO emissions for $E_{\text{NH}_3} = 0\text{--}50\%$ also decrease significantly with increased δ . The emissions for $E_{\text{NH}_3} = 0\%$ are originally less than the others, and decrease with increasing δ . For $E_{\text{NH}_3} = 100\%$, however, the emission decrease, with respect to increased δ , is small.

Figure 4.10 shows that time histories for NO concentrations for various δ for (a) $E_{\text{NH}_3} = 0\%$, (b) $E_{\text{NH}_3} = 20\%$ and (c) $E_{\text{NH}_3} = 100\%$, respectively. For $E_{\text{NH}_3} = 0\%$ and 20% , the sharp NO increases are completed within ~ 1 ms. The final concentrations for $E_{\text{NH}_3} = 20\%$ are over one thousand times larger than that for $E_{\text{NH}_3} = 0\%$, and it is confirmed that the NO production rates decrease as δ increases. The behavior for $E_{\text{NH}_3} = 100\%$ is completely different, in that the rates of NO production are very slow, and it takes more than 10 ms for NO production to settle. The NO production occurs here in two stages. As δ increases, the production rates become slower in the first stage, and faster in the second stage. Eventually, the NO concentrations show similar values regardless of δ , as can be seen in Fig. 4.9.

In summary, the parameters such as E_{NH_3} , δ , λ , and T affect NO emissions, and, importantly, NO emissions are maximized for $E_{\text{NH}_3} = 20\text{--}40\%$, and NO production rates are suppressed by increasing δ .

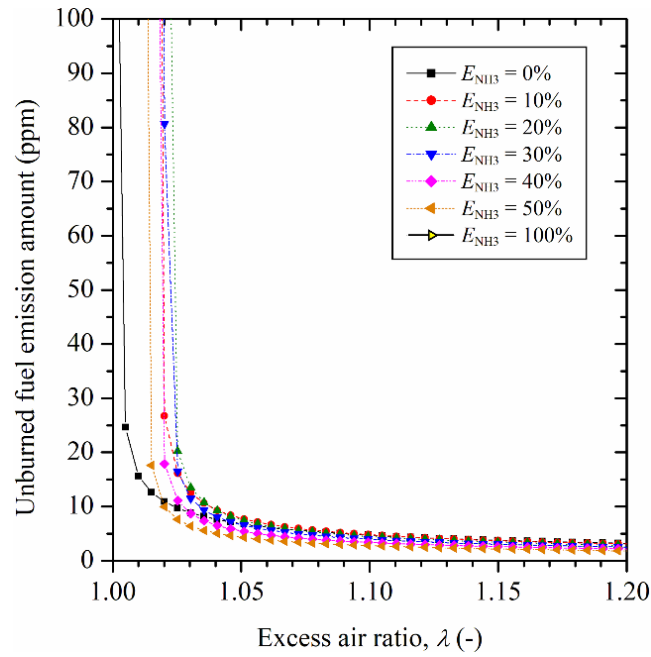


Figure 4.8 Unburned fuel emission amounts after a residence time of 1 s against λ , for various E_{NH_3} , calculated using Okafor-mec. The other parameters are fixed as $\delta = 0\%$, $T = 1250^\circ\text{C}$ and 1 atm. The emissions are corrected at 11% O_2 .

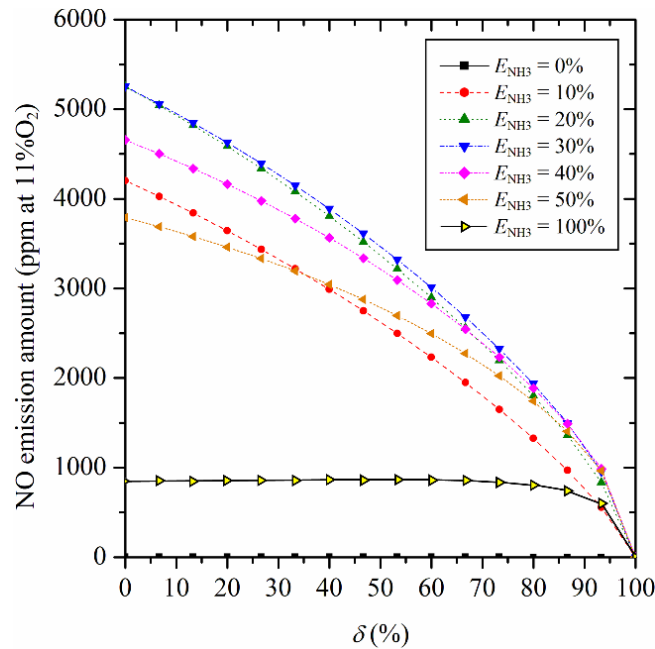


Figure 4.9 NO emission amounts after a residence time of 1 s against δ , for various E_{NH_3} , calculated using Okafor-mec. The other parameters are fixed as $\lambda = 1.10$, $T = 1250^\circ\text{C}$ and 1 atm. The emissions are corrected at 11% O_2 .

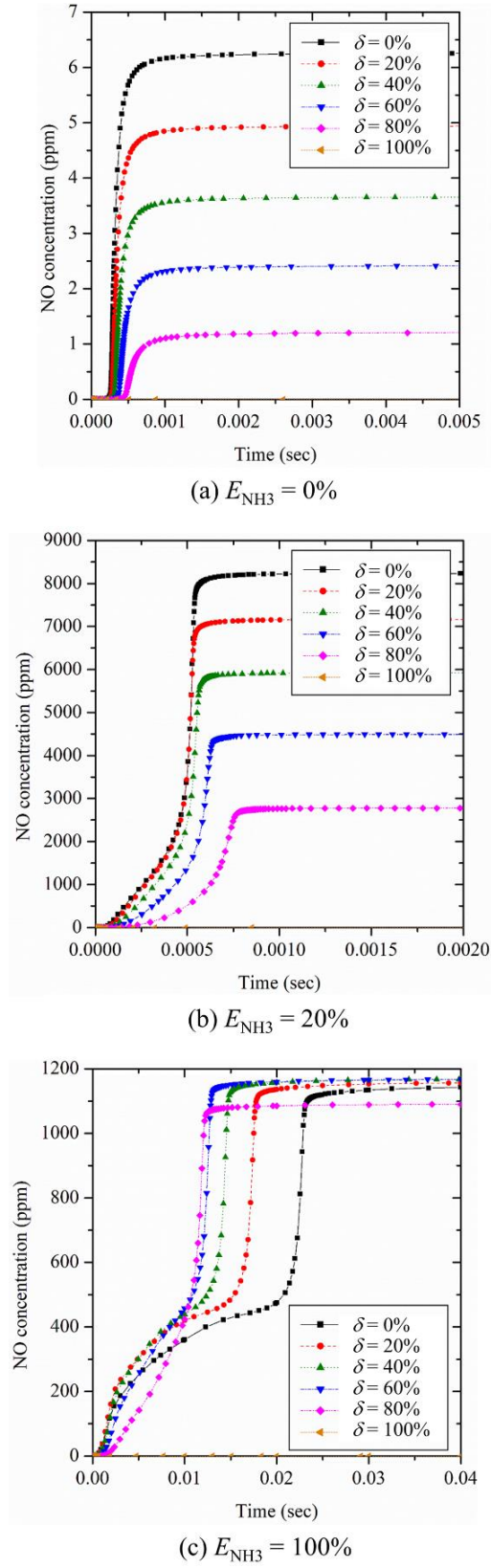


Figure 4.10 Time histories for NO concentrations for various δ for (a) $E_{\text{NH}_3} = 0\%$, (b) $E_{\text{NH}_3} = 20\%$ and (c) $E_{\text{NH}_3} = 100\%$, calculated using Okafor-mec.

4.3.2 Validity of RANS simulations

In this section, for city gas MILD combustion, the validity of our simulation methods is approached by comparing experimental and simulation results. Taking our experimental results, Fig. 4.11 shows that the NO emission amounts measured in our experiments against λ for various L , with D fixed at 50.0 mm. The L denotes the distance between the air nozzles and the fuel nozzle, and the D denotes the inner diameter of the air nozzles. The NO concentrations are corrected at 11% O₂, and the error bars in each plot indicate measurement errors. The NO emissions increase as λ increases, and so, since unburned hydrocarbons are detected when $\lambda < 1.10$, the minimum λ value is set to 1.10. The NO emissions decrease as L increases, becoming 20 ppm or less, at $\lambda = 1.10$ and $L = 190$ mm. Figure 4.12 shows that the NO emission amounts measured experimentally against D for various L , with λ fixed at 1.10. The NO concentrations are corrected at 11% O₂. As D decreases and air injection speed increases, the NO emissions decrease. The NO emissions become less under conditions of large L and small D .

The validity of the simulation method is confirmed with reference to these experimental results. Figure 4.13 shows that the comparison of NO emission amounts with respect to D for the experimental and simulation results. The simulation expresses the same NO emission tendencies as the experiments, in relation to parameters L and D , and the NO emission amounts predict in the simulation are within an error of 6 ppm or less, compared to the experimental results. Figure 4.14 shows that the comparison between the experiment and simulation, for temperatures at each measurement point under conditions of $D = 50.0$ mm, $L = 75.0$ mm, and $D = 19.4$ mm, $L = 190$ mm. The maximum error for all the measurement points is 24°C. Points 1 to 9 indicate temperatures at the position 100 mm below the ceiling, and some points have errors of more than 15°C, although the other errors are within 5°C. Point 11 indicates temperature at the center position in the furnace, and here, the simulation result is 5.6°C higher than the experimental result, under the conditions of $D = 50.0$ mm and $L = 75.0$ mm, and 16°C lower under the conditions of $D = 19.4$ mm and $L = 190$ mm. The temperature prediction errors at point 11 cause the NO emission prediction errors, that is, the simulations overestimate the NO emission amount for $D = 50.0$ mm and $L = 75.0$ mm and

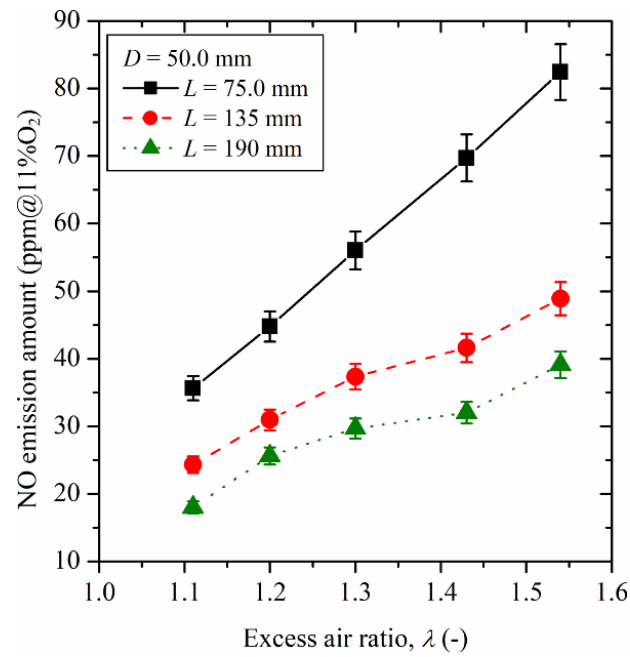


Figure 4.11 NO emission amounts measured in our experiments against λ for various L , with D fixed at 50.0 mm. The NO concentrations are corrected at 11% O₂.

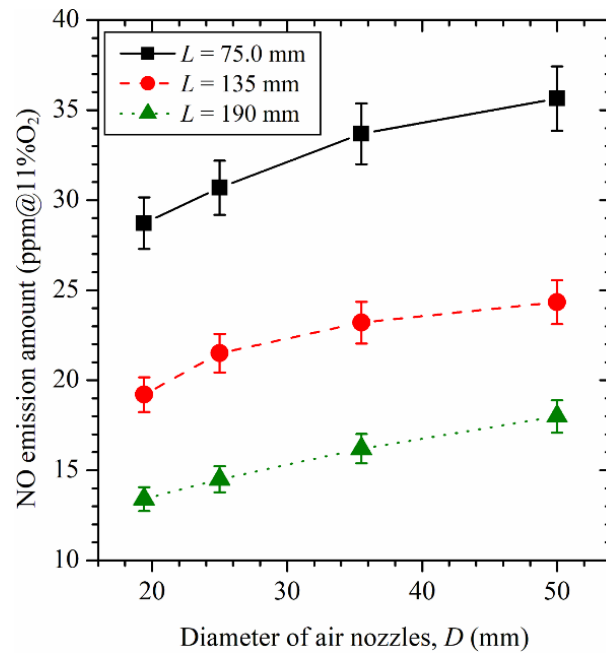


Figure 4.12 NO emission amounts measured experimentally against D for various L , with λ fixed at 1.10. NO concentrations are corrected at 11% O₂.

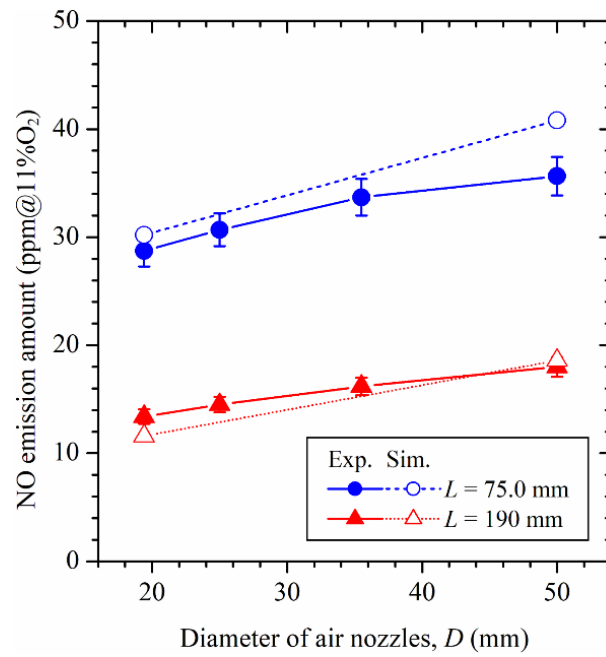


Figure 4.13 Comparison of NO emission amounts with respect to D for the experimental and simulation results.

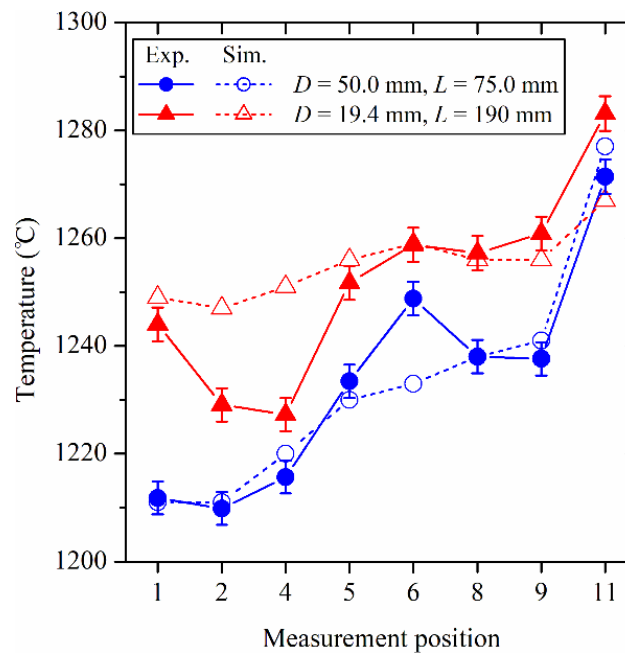


Figure 4.14 Comparison between experiment and simulation for temperatures at each measurement point under conditions of $D = 50.0$ mm, $L = 75.0$ mm, and $D = 19.4$ mm, $L = 190$ mm.

underestimate the amount for $D = 19.4$ mm and $L = 190$ mm.

Based on the above, this simulation method can represent a MILD combustion field for city gas fuel. Furthermore, this method is expanded to ammonia/city gas ($E_{\text{NH}_3} = 20\%$) and pure-ammonia ($E_{\text{NH}_3} = 100\%$) MILD combustion.

4.3.3 Predictions of NO emissions in MILD combustion

By using the simulation methods validated in the previous section, three-dimensional numerical simulations of the MILD combustion fields for $E_{\text{NH}_3} = 20\%$ and 100% are also conducted.

Figure 4.15 shows X-directional velocity distributions in the horizontal cross

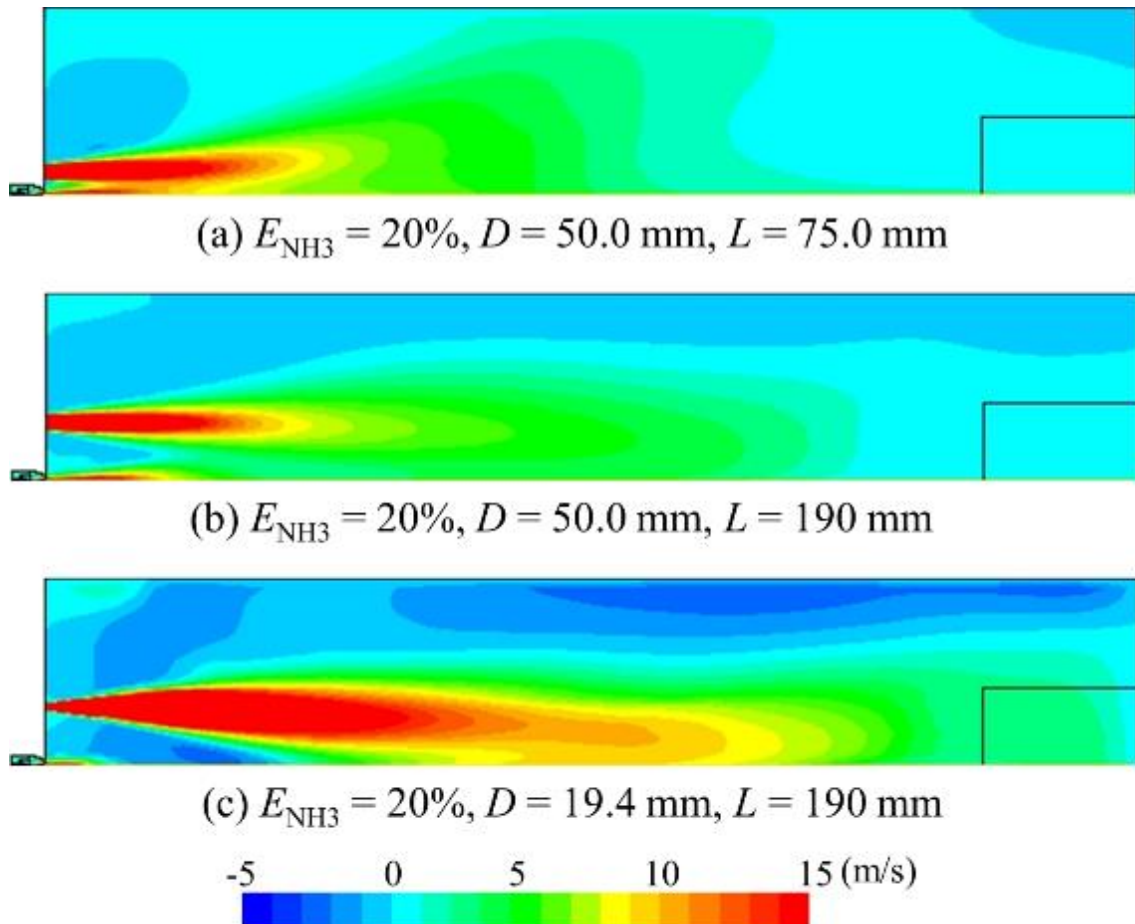


Figure 4.15 X-directional velocity distributions in horizontal cross section at $Z = 0$ for $E_{\text{NH}_3} = 20\%$, under conditions of (a) $D = 50.0$ mm, $L = 75.0$ mm, (b) $D = 50.0$ mm, $L = 190$ mm, and (c) $D = 19.4$ mm, $L = 190$ mm.

section at $Z = 0$, under the conditions of (a) $D = 50.0$ mm, $L = 75.0$ mm, (b) $D = 50.0$ mm, $L = 190$ mm, and (c) $D = 19.4$ mm, $L = 190$ mm, for $E_{\text{NH}_3} = 20\%$. In the condition of $D = 50.0$ mm and $L = 75.0$ mm, the region where the X-directional velocity is negative, is limited, and a small vortex forms just outside the air nozzles. When L increases and in the condition of $D = 50.0$ mm and $L = 190$ mm, a large backflow region forms near the furnace wall. Furthermore, when D decreases in the condition of $D = 19.4$ mm and $L = 190$ mm, the reverse flow velocity near the furnace wall is further enhanced, and a reverse flow region is also generated in the region between the fuel and air nozzles.

Figures 4.16 and 4.17 show the H_2O mole fraction distributions in the horizontal

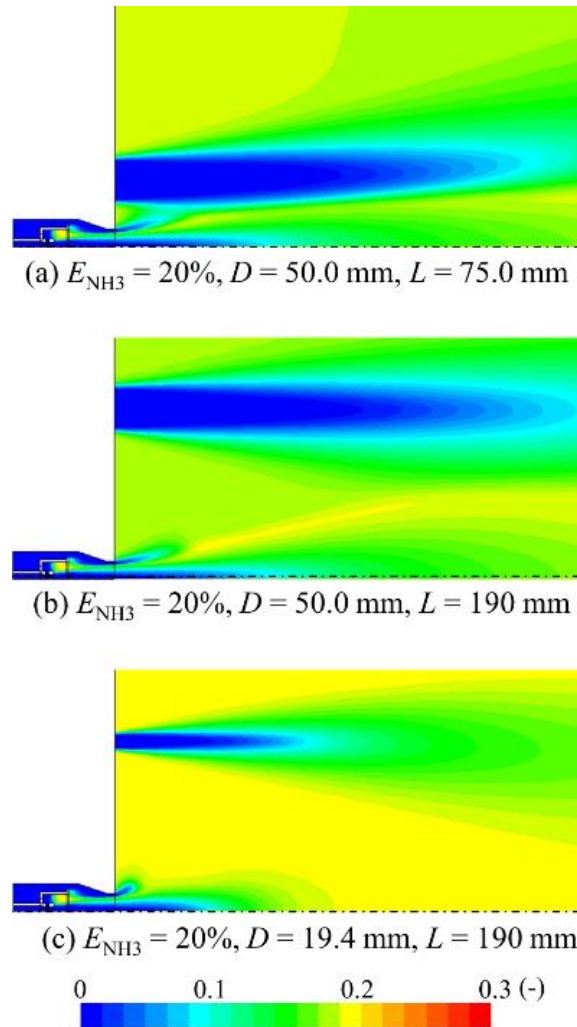


Figure 4.16 H_2O mole fraction distributions in horizontal cross section for $E_{\text{NH}_3} = 20\%$ under conditions of (a) $D = 50.0$ mm, $L = 75.0$ mm, (b) $D = 50.0$ mm, $L = 190$ mm, and (c) $D = 19.4$ mm, $L = 190$ mm.

cross section for various D and L conditions, for $E_{\text{NH}_3} = 20\%$ and 100% . It can be seen that, when $E_{\text{NH}_3} = 20\%$, the H_2O concentration increases in the region between the fuel and the air nozzles, as L increases and D decreases. When $D = 19.4$ mm and $L = 190$ mm, D is more than 90% , when converted from the H_2O concentration to the dilution rate. Even in the condition of $E_{\text{NH}_3} = 100\%$, it is confirmed that the H_2O concentration increases with increased L and decreased D .

Figures 4.18–4.20 show the temperature distributions of the horizontal cross section under various D and L conditions, for $E_{\text{NH}_3} = 0\%$, 20% , and 100% . For $E_{\text{NH}_3} = 0\%$, the maximum flame temperature exceeds 1800°C in the condition of $D = 50.0$ mm and $L = 75.0$ mm, but this decreases to approximately 1500°C in the condition of $D = 19.4$ mm and $L = 190$ mm. The MILD combustion is promoted, as the result, the maximum temperature decreases and the ambient temperature inside increases. This behavior is also confirmed by the results shown in Fig. 4.14. Even for $E_{\text{NH}_3} = 20\%$, it can be confirmed that the maximum flame temperature decreases with increased L and decreased D , and the ambient temperature inside gradually increases. For $E_{\text{NH}_3} = 100\%$, the temperature

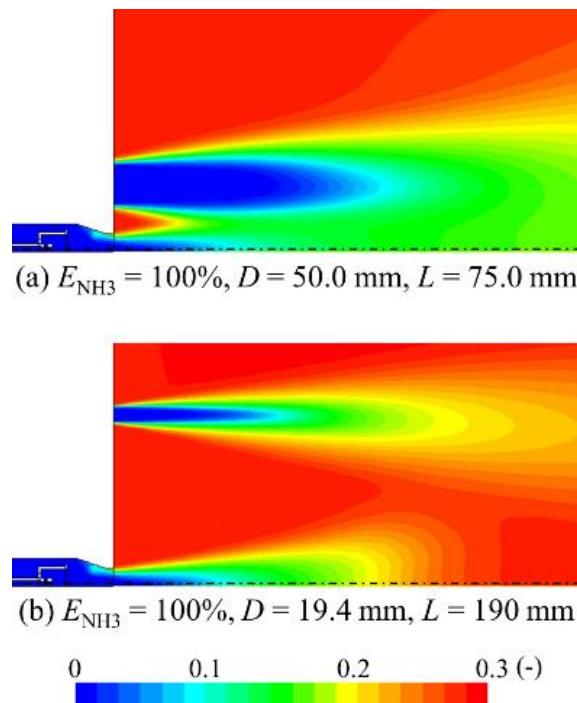


Figure 4.17 H_2O mole fraction distributions in horizontal cross section for $E_{\text{NH}_3} = 100\%$ under conditions of (a) $D = 50.0$ mm, $L = 75.0$ mm, and (b) $D = 19.4$ mm, $L = 190$ mm.

decreases as a whole, so the temperature peak is not observed in the conditions of both $D = 50.0$ mm, $L = 75.0$ mm, and $D = 19.4$ mm, $L = 190$ mm. For $E_{\text{NH}_3} = 0\%$ and 20% , a stable flame forms in the pilot burner and the temperature rises, however, for $E_{\text{NH}_3} = 100\%$, burner temperature rise is not seen and flame extinction occurs. It should be noted, however, that this simulation method is not fully verified, in terms of whether or not the phenomenon of pilot burner ignition and extinction can be simulated.

Fig. 4.21 shows that the NO emission amounts predicted by the simulations for $E_{\text{NH}_3} = 0\%$, 20% , and 100% , in the conditions of $D = 50.0$ mm, $L = 75.0$ mm, and $D = 19.4$ mm, $L = 190$ mm. Regardless of E_{NH_3} , the NO emission amounts decrease with increased L and decreased D . For $E_{\text{NH}_3} = 0\%$ and 100% , the NO emission amounts can be reduced to 100 ppm or less by promoting MILD combustion. The D and L are very important parameters for reducing NO emissions, regardless of E_{NH_3} . For $E_{\text{NH}_3} = 20\%$, a 0.39-fold reduction in D and a 2.5-fold increase in L make the NO emissions 56% and 74%, respectively. In other words, D is a more important design variable. This change in D is replaced by that in the air supply velocity and its Reynolds number, Re . That is, it is

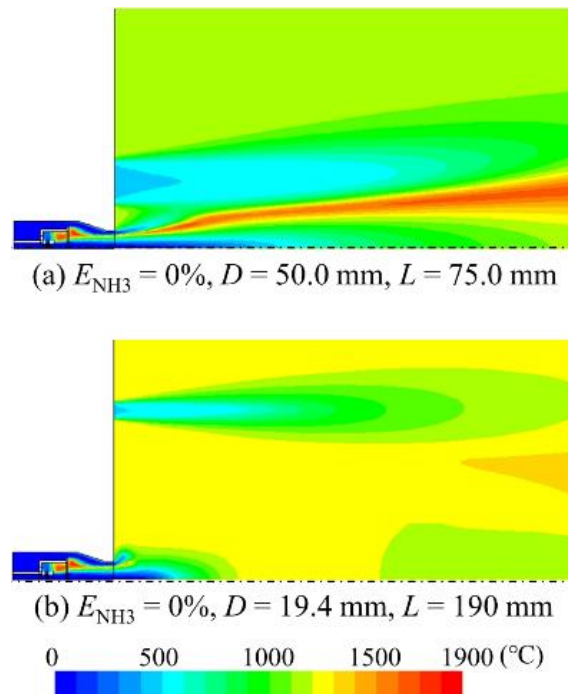


Figure 4.18 Temperature distributions of horizontal cross section for $E_{\text{NH}_3} = 0\%$ under conditions of (a) $D = 50.0$ mm, $L = 75.0$ mm, and (b) $D = 19.4$ mm, $L = 190$ mm.

equivalent to the increase from 22 m/s ($Re \sim 14000$) to 140 m/s ($Re \sim 35000$). For $E_{NH_3} = 20\%$, the NO emission amounts were not sufficiently reduced within the design parameter range examined in our study so that more extensive consideration of other parameters

will be required in future work.

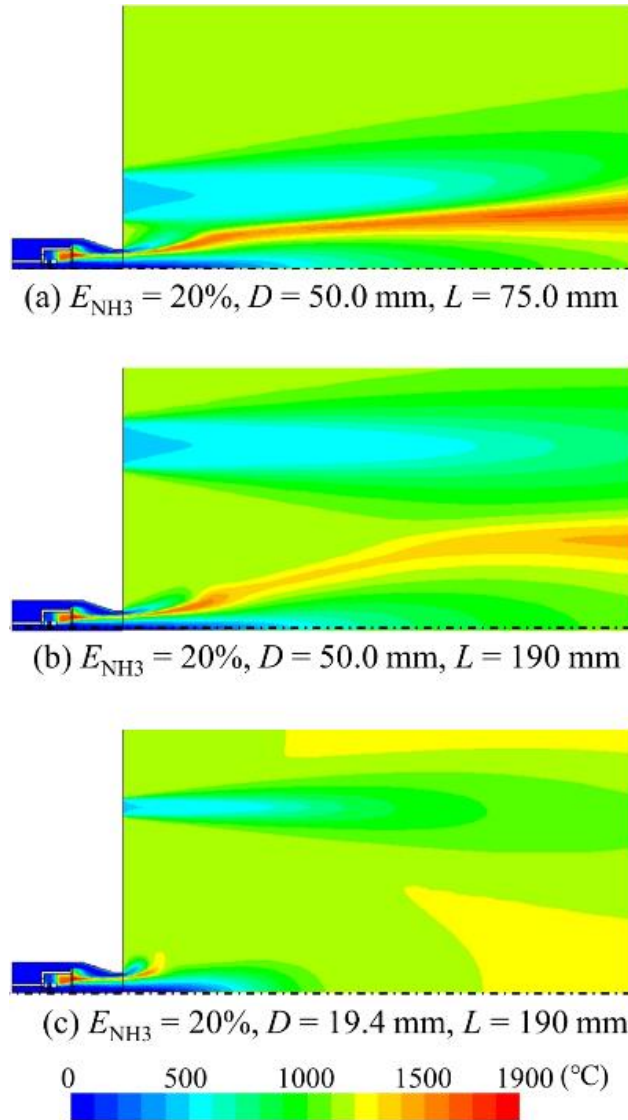


Figure 4.19 Temperature distributions of horizontal cross section for $E_{\text{NH}_3} = 20\%$ under conditions of (a) $D = 50.0$ mm, $L = 75.0$ mm, (b) $D = 50.0$ mm, $L = 190$ mm, and (c) $D = 19.4$ mm, $L = 190$ mm

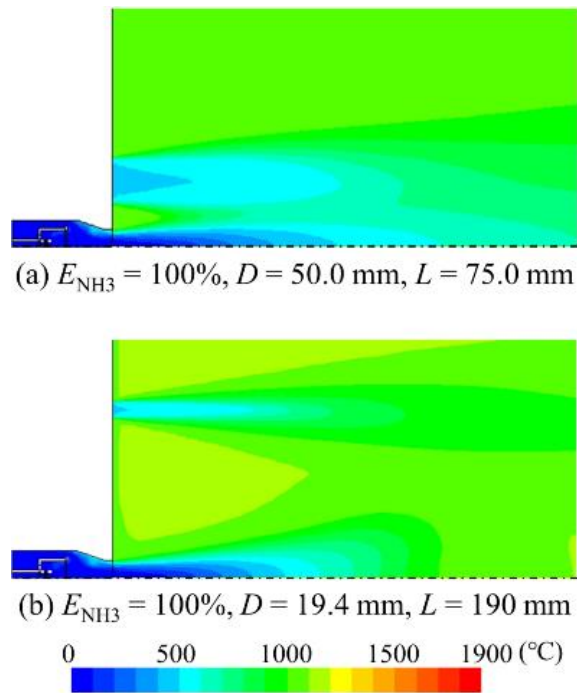


Figure 4.20 Temperature distributions of horizontal cross section for $E_{\text{NH}_3} = 100\%$ under conditions of (a) $D = 50.0$ mm, $L = 75.0$ mm, and (b) $D = 19.4$ mm, $L = 190$ mm.

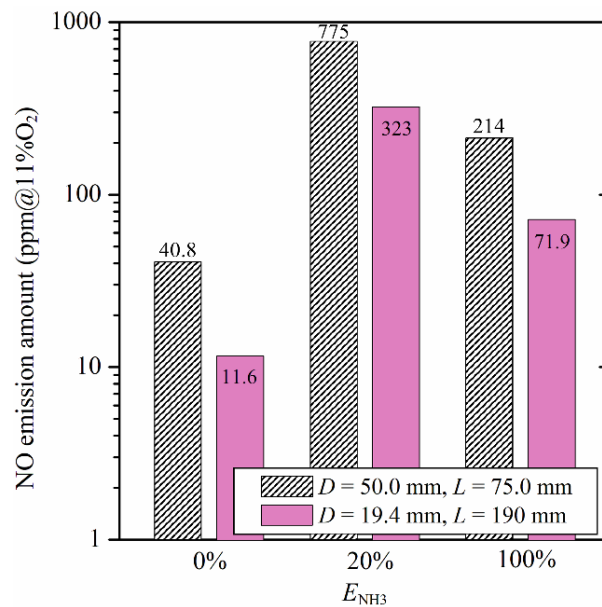


Figure 4.21 NO emission amounts predicted by simulations for $E_{\text{NH}_3} = 0\%$, 20% , and 100% , in the conditions of $D = 50.0$ mm, $L = 75.0$ mm, and $D = 19.4$ mm, $L = 190$ mm.

4.4 Conclusions

In this chapter, mixtures of ammonia and city gas were used as fuels, and the effectiveness of MILD combustion and changing ammonia concentration in the fuels on NO emissions reduction was investigated, using zero-dimensional simulations and three-dimensional RANS simulations. The obtained findings were as follows.

- (1) The zero-dimensional kinetic simulations show that NO emissions are the highest at $E_{\text{NH}_3} = 20\text{--}40\%$, and dilution of the fuel and oxidizer due to EGR reduces the NO emissions, regardless of the E_{NH_3} value. Furthermore, the three-dimensional RANS simulations of an actual furnace revealed that MILD combustion is effective for reducing NO emission amounts, regardless of the E_{NH_3} variation.
- (2) The three-dimensional RANS simulations suggest that for burner design, the oxidizer nozzle diameter D and distance between fuel and oxidizer nozzles L are important parameters, with NO emissions reduced by decreasing D and increasing L .

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Chapter 5: Conclusions

5.1 Summary and conclusions

In this study, the goal was to construct the numerical simulation methods which can simulate for realistic combustors and predict their NO and CO emissions. In this thesis three combustion fields were targeted: the first is the water sprayed combustion for SCV burners which have water spray systems for suppressing NO production (Chapter 2), the second is ammonia/methane premixed swirl combustion for a laboratory scale gas turbine combustor (Chapter 3), and the third is Moderate or Intense Low-oxygen Dilution (MILD) combustion of ammonia/city gas (Chapter 4).

In Chapter 2, the water sprayed combustion in the SCV burners was focused on. This chapter had three purposes. The first purpose was to develop a large-scale SCV burner and to investigate the characteristics of the NO and CO emissions in detail. LESs/5D-NA-FPV were performed and the validity is assessed by comparing with the experiments. The second purpose was to investigate the emission prediction accuracy of the LES/5D-NA-FGM and the LES/5D-NA-FPV. The LESs employing two kinds of different flamelet approaches were performed for a medium-scale SCV burner. The third was to develop a neural network model based on the 5D-NA-FGM in order to reduce the library sizes. The neural network is learned by using the library of the 5D-NA-FGM as a teacher dataset. The LESs employing the neural network model were performed, and for comparison, the combustion experiments using burner equipped with a water spray nozzle were also conducted. In this study, we found new findings such as:

- (1) The development of a large-scale SCV burner has been successful. The NO emissions decreased as the water spray amount increase and record less than 50 ppm corrected at $O_2 = 5\%$ regardless of the loads. The CO emissions increase as the water spray amount increased, but remained at 120 ppm maximum.
- (2) The water-sprayed combustion is generally reproduced by LES/5D-NA-FPV and LES/5D-NA-FGM for large-scale and medium-scale SCV burners. Both can account for the latent heat loss, which lowers the flame temperature and suppresses the combustion reactions, and qualitatively capture the trends of the effects of the water

spray flow rate on the NO and CO emissions, that is, they decrease and increase with increasing water spray rate, respectively. This suggests that the simulation methods can be a powerful tool for the optimal design of SCV burners.

- (3) For the medium-scale SCV burner, the NO and CO emissions predicted by LES/5D-NA-FGM agree much better with those of the experiments than those of LES/5D-NA-FPV. This is because the fuel and air are mixed well around the burner plate, and the characteristics of premixed flames become evident.
- (4) The neural network model based on the 5D-NA-FGM is in good agreement with the original flamelet library for both the temperature and production rates of the progress variable as well as the CO concentrations and the reaction rates of NO. However, further improvements are required to accurately predict CO and NO concentrations with local peaks for the control variables.
- (5) The temperature trends for both LES results using the neural network model and the conventional technique can be generally captured in the experiment in the absence of water spray. In the condition with water spray, the water evaporation causes the temperature decrease, CO increase, and NO production suppression, as in the experiment. For a quantitative evaluation, it is necessary to improve the accuracy of the neural network model and boundary conditions after examining the validity of the flamelet method.

In Chapter 3, the ammonia/methane premixed swirl combustion in a laboratory scale swirl combustor were focused on. The purpose was to predict NO and CO emissions through the LESs/NA-FGM. LESs/FGM were also conducted for comparison. Also, the reaction characteristics of NO and CO in ammonia/methane combustion were also investigated through zero-dimensional calculation using the detailed chemical reaction mechanism. In this study, we clarified:

- (1) The NO and CO emissions predicted by the LES/NA-FGM agree well with the experiments much better than the LES/FGM. This is because the NO and CO reactions for ammonia/methane/air combustion are quite susceptible to H and OH radicals' concentrations and gas temperature. This suggests that it is essential to take into account the effects of various heat losses caused by radiation and cold walls in predicting the NO and CO emissions for the combustion of ammonia as a primary

fuel.

- (2) The NO emissions are successfully predicted using the LES/NA-FGM for $\phi > 1.04$, whereas it is overestimated at $\phi = 0.840$. This is considered to be due to the UCSD-mech used for generating the flamelet libraries. This mechanism tends to overestimate S_L , which may produce NO excessively in a flame zone.

In Chapter 4, the ammonia/city gas MILD combustion in a high temperature air furnace was focused on. The purpose is to investigate the NO emission characteristics for ammonia/city gas MILD combustion, and to find important burner design parameters for the reduction of NO emissions using zero-dimensional simulations and three-dimensional RANS simulations. In this study, we showed:

- (1) From the zero-dimensional kinetic simulations, the NO emission amounts are the highest for $E_{\text{NH}_3} = 20\text{--}40\%$, and the dilution of the fuel and oxidizer by EGR reduce the NO emissions, regardless of E_{NH_3} value. In addition, the three-dimensional numerical simulations of an actual furnace show that MILD combustion is effective for reducing NO emissions regardless of the E_{NH_3} variation.
- (2) The three-dimensional simulations suggest that for burner design, D and L are important parameters, with NO emissions reduced by decreasing D and increasing L .

Above results can lead the conclusion that LES/5D-FGM and LES/NA-FPV are valid methods to design SCV burners with water spray system to reduce NO and CO emissions, and LES/NA-FGM can quantitatively predicts NO and CO emissions in ammonia/methane premixed swirl combustion, especially at fuel-rich conditions. Also, RANS simulations employing EDC model can predict NO emissions for MILD combustion.

5.2 Suggestions for possible future research

There are some possible future extensions of the present work that could provide valuable information for predicting NO and CO emissions more accurately. In particular, the non-adiabatic flamelet approaches (5D-NA-FGM and 5D-NA-FPV) for water sprayed combustion were newly developed, and the NA-FGM was also applied to

ammonia/methane premixed swirl combustion for the first time. Therefore, issues and future work regarding the non-adiabatic flamelet approaches are addressed.

- (1) In this thesis, two types of flamelet methods (FGM and FPV) were made non-adiabatic. The both are based on pure-premixed flames or pure-diffusion flames. The combustion state in industrial combustors is complicated, and premixed flame and diffusion flame may coexist. In this case, the 5D-NA-FGM and 5D-NA-FPV cannot completely express the combustion state in the entire region of the combustion chamber. In Chapter 2, neither the LES/5D-NA-FGM or the LES/5D-NA-FPV could quantitatively predict NO and CO emissions. Of course, it is difficult to quantitatively evaluate NO and CO emissions in actual combustors, which are large in size, complicated in shape and flow field and often the boundary conditions cannot be grasped in detail. Needless to mention, it is important to be able to obtain a guideline for optimal design even if they cannot be evaluated quantitatively. However, in order to realize the concept of the digital twin in the future, NO and CO emissions must be quantitatively predicted through simulations. As a further extension of the 5D-NA-FGM and 5D-NA-FPV proposed in this study, for example, non-premixed and premixed flamelets by Hu and Kurose [1] can be addressed. They combined a diffusion flamelet and a premixed flamelet, and LESs employing the combined flamelets were performed for spray flames. By using this model for my combustion field, it may be expected to express both the propagations the characteristics of the premixed flame and the ignition/extinction/lifting as the characteristics of the diffusion flame.
- (2) Another issue is to reduce the data size of the flamelet library in order to extend the range of non-adiabatic flamelet methods. The neural network modeling tried in Chapter 2 has a very high potential as a data compression method. However, this method has three difficulties. The first is to select the appropriate detailed chemical reaction mechanism to generate the library needed to represent complex combustion fields. In particular, the development of the reaction mechanism about ammonia combustion discussed in Chapter 3 is still now under development. In addition, the use of syngas and biogas may increase to reduce CO₂ emissions, and reaction mechanisms for these or a mixed fuel of these and carbon-free fuel (like ammonia) will be required. The second is to take time to prepare the huge amount of the results

of one-dimensional calculations used as teacher data. In Chapter 2, the five-dimensional flamelet library was developed, but the sum of data size of the one-dimensional calculation results, which are used as teacher data, was 35 GB. Third, there is no established method for multifaceted accuracy verification of the generated neural network model. If the same solution as the theoretically calculated one-dimensional calculation result is pursued, over-learning occurs. The one-dimensional calculation results have "sparseness" in certain areas for each control variable. There is a risk of taking non-physical values in the area. However, if these problems can be solved, the neural network modeling can be used in engineering. It will enhance the design and efficiency of industrial burners.

- [1] Y. Hu and R. Kurose, "Nonpremixed and premixed flamelets LES of partially premixed spray flames using a two-phase transport equation of progress variable," *Combustion and Flame*, vol. 188, pp. 227-242, 2018, doi: 10.1016/j.combustflame.2017.10.004.