Numerical Study of NO_x Emission in High Temperature Air Combustion*

Hongsheng GUO**, Yiguang JU***,
Kaoru MARUTA**, Takashi NIIOKA**
and Jun'ichi SATO****

The characteristics of NO_x emission in the high-temperature air combustion technology were investigated numerically. Two kinds of methods were used. Detailed chemistry and transport properties were used for a two-dimensional laminar diffusion flame, and a simplified reaction scheme was used for calculating a turbulent flame in a furnace. Results show that the thermal mechanism dominates NO_x emission in high-temperature air combustion. If only the preheated air temperature is increased, NO_x emission will rise significantly. However, the combination of air preheating and flue gas recirculation not only improves the combustion efficiency, but also suppresses NO_x emission in the combustion process.

Key Words: Combustion, NO_x, Simulation, Furnace, Flue Gas

1. Introduction

The world's basic and advanced-technology industries rely heavily on an adequate supply of high-quality energy, the production of which closely depends on the combustion technology. The international competitiveness between these industries hinges on their ability to use more efficiently and cleanly, through advanced combustion technology, available fuel resources. The design of advanced combustion systems and the process for the efficient and clean use of these fuels can be greatly enhanced by a clear understanding of the combustion and pollutant emission mechanisms, and the effects of various parameters on them.

The high-temperature air combustion technology, which was developed by Tanaka et al.⁽¹⁾, has had a significant impact on the design and development of advanced industrial furnaces for energy saving, furnace downsizing and pollutant reduction. Generally, preheating the inlet air will lead to an increase in the temperature in the furnace which will greatly increase NO_x emission, although the combustion efficiency and the heat transfer process in the furnace can be improved. However, the combination of preheating air and the flue gas recirculation, which was defined as high-temperature air combustion technology, not only improves the combustion efficiency but also decreases NO_x emission.

In this study an understanding of the combustion and NO_x emission mechanisms and the effects of various parameters on them in high-temperature air combustion technology through numerical calculations has been focused on.

In recent years, NO_x emission in the hydrocarbon fuel combustion process was investigated by many researchers both experimentally and numerically⁽²⁾⁻⁽¹⁰⁾. There are two kinds of strategies used in the numerical simulation of NO_x emission.

* Received 12th December, 1996

** Institute of Fluid Science, Tohoku University, Katahira, Aoba-ku, Sendai 980-8577, Japan

*** Department of Aeronautics and Space Engineering, Tohoku University, Aramaki Aoba, Aoba-ku, Sendai 980-8579, Japan

**** Research Institute, Ishikawajima-Harima Heavy Industry Co., Ltd., Toyosu, Koto-ku, Tokyo 135-0061, Japan

One is the calculation of NO_x emission by detailed chemistry and transport properties, and the other is the calculation by some simplified reaction mechanisms. The former method provides a clear understanding of the NO_x emission mechanism. However, any attempt to simulate all the corresponding intermediate reactions in practical furnaces would precipitate a computational catastrophe, because more than 200 intermediate species have been identified in the combustion of the typical hydrocarbon fuels⁽²⁾. Thus this kind of method can only be used for some simpler laminar flames^{(3)–(6)}. The latter method can be used for some practical turbulent combustion processes^{(7)–(10)}, but the results are closely dependent on the simplified reaction scheme. Unfortunately there does not exist such a good simplified scheme which can reproduce the results obtained from the detailed chemistry in any case. Therefore, the simplified reaction scheme can only be used for a qualitatively understanding of the effects of various parameters on NO_x emission in practical turbulent combustion.

Both of the above methods were used in the present study. A simplified reaction scheme was used for investigating the effects of various parameters on NO_x emission and combustion in a two-dimensional turbulent flame, and the detailed chemistry and transport properties were used for the modeling of NO_x emission of a two-dimensional laminar diffusion flame to clearly understand the NO_x emission mechanisms in high-temperature air combustion. Special attention was paid to the effects of preheated air temperature and the fraction of flue gas mixed into the air on NO_x emission and combustion. In addition, the effect of the Lewis number on the NO_x emission in the furnace was also checked numerically.

2. Numerical Model

2.1 Calculation with a simplified reaction scheme for turbulent combustion in a furnace

The furnace we modeled is a two-dimensional chamber of size 1.8 × 4.0 m², and is shown in Fig. 1(A). The fuel used in the calculation is methane.

The flow field and concentrations of various species were obtained by solving the Reynolds-averaged conservation equations using the SIMPLE algorithm⁽¹¹⁾. Turbulence closure was obtained using the *k*- ϵ model⁽¹²⁾.

The combustion model employed for methane oxidization in the present investigation used a one-step reaction mechanism⁽¹³⁾. The thermal NO formation rate used in the calculation is the one suggested by Kenbar et al.⁽⁷⁾, based on the extended Zeldovich mechanism. The quantity of the prompt-NO formed

Table 1 Calculation conditions, where *f* is the fraction of CO₂ mixed into the inlet air, *T*_{air} is the inlet air stream temperature, *T*_f is the inlet fuel temperature, *V*_{air} is the inlet air stream velocity, *V*_f is the inlet fuel velocity and *Le* is the Lewis number

Case	<i>f</i>	<i>T</i> _{air} (K)	<i>T</i> _f (K)	<i>V</i> _{air} (m/s)	<i>V</i> _f (m/s)	<i>Le</i>
1	0.3	1500	300	128.00	75.0	1.0
2	0.3	1300	300	110.93	75.0	1.0
3	0.3	1000	300	85.33	75.0	1.0
4	0.2	1500	300	116.30	75.0	1.0
5	0.2	1300	300	100.80	75.0	1.0
6	0.2	1000	300	77.54	75.0	1.0
7	0.1	1500	300	107.25	75.0	1.0
8	0.1	1300	300	92.95	75.0	1.0
9	0.1	1000	300	71.50	75.0	1.0
10	0.5	1500	300	165.50	75.0	1.0
11	0.0	1500	300	100.00	75.00	1.0
12	0.0	1300	300	86.67	75.0	1.0

in practical combustion systems is small compared to the total NO formed. In the present investigation the global kinetic parameter, recommended by Missaghi et al.⁽¹⁴⁾, was employed to predict the prompt-NO emission.

The enthalpies of the species were obtained from JANAF thermochemical tables. Since we are only concerned with the qualitative effects of various parameters on the NO_x emission in high-temperature air combustion, the chemistry/turbulence interactions were neglected in the calculation. NO_x emission and methane oxidization were calculated in a full coupling way. Unless otherwise indicated, the turbulent Prandtl number was taken as 0.9 and the Lewis number was taken as 1.0. CO₂ was used as the flue gas in the calculation for the sake of simplicity.

The calculation conditions for different cases are given in Table 1.

For comparing the combustion characteristics, we define the combustion efficiency η as:

$$\eta = \frac{m_{f,in} - m_{f,out}}{m_{f,in}} \times 100 \% \quad (1)$$

Where $m_{f,in}$ and $m_{f,out}$ are the inlet and outlet fuel mass flow rates, respectively.

2.2 Calculation with the detailed chemistry for laminar jet flame

A diffusion flame of methane injected through a nozzle into an infinite air stream is considered, as shown in Fig. 1(B). The inlet velocities of the methane and air streams are, respectively, 50 cm/s and 30 cm/s. The inlet fuel temperature is specified at 300 K while the air temperature is raised from 300 K to 1300 K. The initial pressure is 1 atm. The Reynolds

(A) Two-dimensional furnace for calculation with simplified reaction scheme

(B) Two-dimensional laminar jet diffusion flame for calculation with detailed chemistry

Fig. 1 Schematic illustration of the computational domain; (A) Furnace, (B) Laminar jet flame

number is lower than 300, and thus the flow can be assumed to be laminar. Chemistry mechanism used in this study is basically the one presented by Miller and Bowman⁽²⁾. The data obtained for the $\text{H} + \text{O}_2 = \text{OH} + \text{O}$ reaction by Yu et al.⁽¹⁵⁾ was used. In addition, the reaction $\text{CH}_3 + \text{CH}_3 = \text{C}_2\text{H}_4 + \text{H}_2$ was added to the C_2 chemistry for high-temperature air combustion consideration. Transport coefficients and thermodynamic properties were calculated using the CHEMKIN package⁽¹⁶⁾. Governing equations were discretized using a fourth-order upwind scheme and were solved by a revised version of PISO⁽¹⁷⁾. The computational domain is a $5 \text{ cm} \times 4.5 \text{ cm}$ rectangle with 131×121 grid points.

In order to quantitatively evaluate NO_x emission for a two-dimensional laminar flame, the emission index is defined as:

$$EI_{\text{NO}_x} = \frac{\iint \omega_{\text{NO}_x} dx dy}{-\iint \omega_{\text{CH}_4} dx dy} \times 1000 \text{ g/kg} \quad (2)$$

Where ω_{NO_x} and ω_{CH_4} are the production and consumption rates of NO_x and CH_4 , respectively.

3. Results and Discussions

The preheated air temperature and fraction of flue gas mixed into the air are two key parameters which affect the combustion efficiency and NO_x emission in the high-temperature air combustion technology.

We investigated the effects of these parameters by both methods. In addition, the effect of the Lewis number on NO_x emission in the furnace was investigated.

3.1 The effect of preheated air temperature

By maintaining constant fuel temperature and velocity at the furnace inlet, we investigated the effect of preheated air temperature when the excess air was kept at 25%. Figures 2 A and B (cases 1 and 2 in Table 1) show the temperature distributions for two typical inlet air temperatures (1500 K and 1300 K, respectively) with all other conditions the same. It is obvious that with the increase of preheated air temperature, the temperature in the furnace rises if other conditions are kept constant. When the temperature of the preheated air is 1300 K, the maximum flame temperature in the furnace is 2370 K while it is 2560 K for the preheated air temperature of 1500 K if the fraction of CO_2 mixed into the air was kept at 30%. As a result, the NO concentration in the furnace for the preheated air of temperature 1500 K is much higher than that for the preheated air of temperature 1300 K, as shown in Figs. 3 A and B (corresponding to Figs. 2 A and B, respectively), although the combustion efficiency for the preheated air of temperature 1500 K is higher than that for the preheated air of temperature 1300 K. The NO concentration at the exit for the preheated air of temperature 1300 K is only 69.8 ppm while it is 431.0 ppm for the preheated air of temperature 1500 K. Figure 4 shows the results for the laminar diffusion flame obtained by detailed chemistry and transport properties. Similarly we can find that NO_x emission significantly increases with the rise of the preheated air temperature. In addition, Fig. 4 clearly shows that, at low air temperatures, NO is mainly produced within the reaction zone while at high air temperatures the NO production zone thickens and extends to the air side.

Both Figs. 3 and 4 confirm that the areas of highest NO_x concentration occur within or near the reaction regions. This indicates that NO_x emission rates are highest in the high-temperature zones. Therefore, for the high-temperature air combustion NO_x emission is suggested to be dominated by a thermal mechanism.

Figure 5 shows a comparison of NO_x emission indexes, defined as in Eq. (2), calculated by using the full chemistry and only the Zeldovich mechanism for the laminar diffusion flame. At low temperatures, NO_x emission calculated by the Zeldovich mechanism is much lower than that given by the full chemistry. At temperatures near 900 K, the Zeldovich mechanism yields an equivalent result to that of the full chemistry. At high temperatures, however, Fig. 5 clearly

Fig. 2 Temperature distributions in the furnace, where f is the fraction of CO_2 mixed into the inlet air

Fig. 3 NO_x mass fraction distribution in the furnace, where f is the fraction of CO_2 mixed into the inlet air

shows that the Zeldovich mechanism far overpredicts the results obtained from the full chemistry. The reason is that the prompt NO_x is strongly coupled with the Zeldovich mechanism⁽⁴⁾. Therefore, we can

Fig. 4 NO_x distribution in the laminar jet flame

Fig. 5 Comparison of NO_x emission index between the results obtained from the full chemistry and those obtained from only the Zeldovich mechanism

conclude that the thermal mechanism dominates the NO_x emission in high-temperature air combustion. If only the preheated air temperature is raised, keeping other conditions constant, NO_x emission will significantly increase.

Also, it should be noted that the simplified reaction scheme employed in the present calculation can qualitatively represent the fundamental characteristics of NO_x emission in the high temperature air

Fig. 6 Variations of NO_x concentration at the furnace exit with the inlet air temperature and the fraction (f) of CO_2 mixed into the inlet air

Fig. 7 Variations of NO_x emission index with the inlet air temperature and the O_2 concentration (X_{O_2}) in the inlet air for the laminar jet flame

combustion although the quantitative discussion may not be adequate.

3.2 The effect of flue gas recirculation

In the high-temperature air combustion technology, preheating of air is combined with flue gas recirculation. Keeping the mass flow rate and temperature of the inlet fuel and excess air constant, we conducted a series of calculations to investigate the effect of a fraction of CO_2 mixed into the air on the NO_x emission in the furnace. Figure 6 shows the results (cases 1 - 10 in Table 1). Please note that in order to keep the excess air constant, the inlet velocity of the air stream (the mixture of air and CO_2) changes for different cases. Due to the mixing of CO_2 into the inlet air, the concentrations of O_2 and N_2 in the air stream change for different cases. We can find that NO_x emission can be significantly decreased by mixing CO_2 into the air for the same preheated air temperature.

Fig. 8 Variations of the combustion efficiency with the inlet air temperature and the fraction of CO_2 mixed into the air

Fig. 9 Turbulent kinetic energy distributions in the furnace, where f is the fraction of CO_2 mixed into the inlet air

Particularly for higher air temperatures, the reduction of NO_x emission is considerable. Figure 7 shows the results from the calculations by detailed chemistry and transport properties for a laminar jet flame. Similar to the results obtained from the furnace calculation, with the decrease of O_2 concentration in the inlet air (same as the increase of the CO_2 fraction mixed into the air), NO_x emission decreases if the other conditions are kept the same.

For analyzing the NO_x emission reduction mechanism when mixing CO_2 with air, we check the temperature distributions in the furnace for two typical different cases. Figures 2 A and C show two temperature distributions for the fraction of CO_2 mixed with the inlet air being 30% and 10% (cases 1 and 7 in

Table 1), respectively, while the preheated air temperature is 1 500 K. We find that the temperature drops when the fraction of CO₂ mixed with the air increases. The maximum flame temperature in the furnace is 2 560 K for case 1 of Table 1 (Fig. 2 A), while the maximum flame temperature in the furnace is 2 810 K for case 7 (Fig. 2 C). This maximum flame temperature drop is due to the increase of heat capacity and the intensifying of the mixing process in the furnace (as will be analyzed later). As discussed above, the thermal mechanism dominates the NO_x emission in high-temperature air combustion. Therefore this maximum flame temperature drop leads to the reduction of NO_x emission. Thus, the mechanism for NO reduction by mixing flue gas into the inlet air is due to the flame temperature decrease.

How will mixing CO₂ into the inlet affect the combustion efficiency of the furnace? Our calculation for the furnace shows that mixing CO₂ into the air can improve the combustion efficiency of the furnace when other conditions were kept constant in the high-temperature air combustion technology. Figure 8 shows the variation of combustion efficiency with the inlet air temperature and the fraction of CO₂ mixed with the inlet air. It can be found that the increase in the fraction of CO₂ mixed with the air leads to the rise of the combustion efficiency, other conditions remaining unchanged. This is due to the intensifying of the mixing process in the furnace. Figure 9 shows the distributions of turbulent kinetic energy in the furnace for cases 1 and 7 in Table 1 (corresponding to Figs. 2 A and C, respectively). We find that the turbulent kinetic energy of case 1 is higher than that of case 7, because the inlet velocity of the air stream (mixture of air and CO₂) of case 1 (Fig. 2 A) is higher than that of case 7 (Fig. 2 C) for maintaining the same amount of excess air while keeping the inlet fuel velocity the same for both the cases. As a result, the mixing process between fuel and air in the furnace is intensified. Since the active energy of methane oxidization (48 Kcal/mole) is much lower than that of thermal NO emission (76 Kcal/mole), the key factor affecting the methane oxidization is the mixing process between the air stream and fuel stream, which is different from the thermal NO emission. Therefore the intensifying of the mixing process in the furnace leads to the increase of fuel oxidization rate. Thus mixing CO₂ into the preheated air can improve the combustion efficiency although the temperature in the furnace decreases by a small amount, the other conditions remaining unchanged. On the other hand, since the thermal mechanism dominates the NO mechanism, the key factor affecting NO emission is temperature and therefore the NO emission rate decreases

Table 2 Comparison of the results for two different Lewis numbers, where η is the combustion efficiency, C_{NO} is the NO concentration at the exit of the furnace and T_{max} is the maximum flame temperature in the furnace

Tair(K)	Le	η (%)	C_{NO} (ppm)	Tmax(K)
1300	1.0	81.15	2890.7	2770
1500	1.8	88.18	2530.0	2530

when CO₂ is mixed with the preheated air.

Therefore the combination of air preheating and flue gas recirculation not only improves the combustion efficiency but also decreases the NO_x emission. We can compare the two cases in Fig. 6 (cases 8 and 1 in Table 1), 1 300 K, 10% and 1 500 K, 30% for the preheated air temperature and the fraction of CO₂ mixed into the air, respectively. The combustion efficiency and NO_x concentration at the exit of furnace are 84.07% and 1124.2 ppm for case 8 while they are 90.89% and 431.03 ppm, respectively, for case 1 of Table 1.

3.3 The effect of Lewis number

From the above discussion, we know that the drop of the maximum flame temperature in the furnace results in the reduction of NO emission in the furnace. This stimulates us to check that how the change of the turbulent Lewis number affects the NO emission, as the Lewis number is the ratio of the thermal diffusion coefficient to the mass diffusion coefficient.

Two calculations were conducted to investigate the Lewis number effect on NO_x emission in the practical furnace. In the first calculation (case 12 of Table 1), the preheated air temperature is 1 300 K and the Lewis number is 1.0. The preheated air temperature and the Lewis number were taken as 1 500 K and 1.8, respectively, in the second calculation (case 11). The excess air and the fuel temperature were kept constant and no CO₂ was mixed into the air in both calculations. The results are given in Table 2. We find that the NO_x emission for case 12 is higher than that for case 11 although the preheated air temperature for case 12 is lower. This is because increasing the Lewis number means that the thermal diffusion is faster than the mass diffusion, and thus the temperature distribution in the furnace becomes flatter and the maximum temperature drops. This comparison shows once again that the drop of maximum flame temperature in the furnace results in the reduction of NO emission, even if no flue gas is mixed with the inlet air. Therefore it can be further confirmed that thermal mechanism dominates the NO emission in the high-temperature air combustion technology and the

reduction of NO emission is due to the drop of maximum flame temperature in the furnace.

Note that the above two Lewis numbers are assumed values, because it is very difficult to determine the exact Lewis number for turbulent flow. Usually the turbulent Lewis number is close to unity. Therefore the above calculations were conducted to only to help in the understanding that the thermal mechanism dominates NO emission in the high-temperature air combustion technology and the reduction of NO emission is due to the drop of the maximum temperature in the furnace.

4. Conclusions

Numerical calculations of NO_x emission in high-temperature air combustion were conducted. The effects of the preheated air temperature, the fraction of flue gas mixed into the inlet air and the Lewis number on NO_x emission and combustion in high-temperature air combustion were investigated.

The results show that thermal mechanism dominates NO_x emission in the high-temperature air combustion technology. If only the preheated air temperature is increased, keeping all other conditions the same, the NO_x emission in the furnace will significantly increase although the combustion efficiency can be improved. Mixing flue gas into inlet air leads to the drop of maximum flame temperature in the furnace and thus results in the reduction of NO_x emission. The combination of air preheating and flue gas recirculation not only improves the combustion efficiency but also decreases the NO_x emission in the furnace due to the intensifying of the mixing process and the decrease of the maximum temperature in the furnace.

References

- (1) Tanaka, R., Kishimoto, K. and Hasegawa, T., *Combust. Sci. Technol.*, (in Japanese), Vol. 2, No. 4 (1994), p. 257.
- (2) Miller, J.A. and Bowman, C.T., *Prog. Energy Combust. Sci.*, Vol. 15 (1989), p. 287.
- (3) Hahn, W.A. and Wendt, J.O.L., 18th Symp. (Int.) on Comb., *The Comb. Inst.*, (1981), p. 121.
- (4) Ju, Y., *Trans. Jpn. Soc. Mech. Eng.*, (in Japanese), Vol. 62, No. 599 (1996), p. 314.
- (5) Nishioka, M., Nakagawa, S., Ishikawa, Y. and Takeno, T., *Combust. Flame*, Vol. 98 (1994), p. 127.
- (6) Ju, Y. and Niioka, T., *Combust. Theory and Modeling*, Vol. 1 (1997), p. 243.
- (7) Kenbar, A.M.A., Beltagui, S.A., Ralston, T. and MacCallum, N.R.L., *Combust. Sci. Technol.*, Vol. 93 (1993), p. 173.
- (8) Tomeczek, J., Góral, J. and Garodon, B., 25th Symp. (Int.) on Comb., *The Comb. Inst.*, (1994), p. 1035.
- (9) Boardman, R.D., Eatough, C.N., Germane, G.J. and Smoot, L.D., *Comb. Sci. Technol.*, Vol. 93 (1993), p. 193.
- (10) Al-fawaz, A.D., Dearden, L.M., Hedley, J.T., Missaghi, M., Pourkashanian, M., Williams, A. and Yap, L.T., 25th Symp. (Int.) on Comb., *The Comb. Inst.*, (1994), p. 1027.
- (11) Patanker, S.V., *Numerical Heat Transfer and Fluid Flow*, Hemisphere, Washington, DC., 1980.
- (12) Launder, B.E. and Spalding, D.B., *Comp. Methods Appl. Mech. Eng.*, Vol. 3 (1974), p. 269.
- (13) Westbrook, C.K. and Dryer, F.L., *Combust. Sci. Technol.*, Vol. 27 (1981), p. 31.
- (14) Missaghi, M., Pourhashanian, M., Yap, L.T. and Williams, A., *Proceedings of the American Flame Foundation Conference*, San Francisco, Oct. 1990, IFRF Publications, The Netherlands.
- (15) Yu, C.L., Frenklach, M., Masten, D.A., Hanson, R. K. and Bowman, C.T., *J. of Physical Chemistry*, Vol. 98 (1994), p. 4770.
- (16) Kee, R.J., Dixon-Lewis, G., Warnatz, J., Coltrin, M.E. and Miller, J.A., *Sandia Report*, SAND 86-8246 (1986).
- (17) Issa, R.I., Ahmadi-Befrui, B., Beshay, K.R. and Gosman, A.D., *J. Comput. Phys.*, Vol. 93 (1991), p. 388.