

NUMERICAL STUDY OF REDUCTION OF NO_x EMISSION BY HIGH TEMPERATURE AIR COMBUSTION TECHNOLOGY

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Abstract A numerical study on NO_x emission from an industrial furnace utilizing high temperature air combustion (HTAC) technology was done. The basic concept of the regenerative burner including heating a diluting of fresh air by flue gas was implemented in a two-dimensional furnace model. Governing equations in conjunction with a turbulence model and an overall chemistry model were solved using an implicit numerical scheme. Effect of temperature and O₂ concentration of preheated airflow on NO_x emission from a furnace were investigated. Numerical results show a drastic increase in NO_x emission when temperature or O₂ concentration of preheated airflow increases. High combustion efficiency and low NO_x emission can be achieved when the inlet airflow is preheated up to a high temperature, for instance, about 1500 °K, and O₂ reduced to about 5%.

Key Words Combustion, Industrial Furnace, Numerical Study, NO_x, HTAC

چکیده مطالعه شبیه سازی عددی انتشار NO_x از کوره های صنعتی با استفاده از تکنولوژی HTAC انجام گرفته است. در این جهت با اعمال regenerative burner که شامل پیش گرم هوای رقیق شده است در یک کوره صنعتی دو بعدی فرآیند احتراق شعله دیفیوژن در پاشش جت سوختی متان به داخل جریان هوای با درجه حرارت بالا مورد مطالعه قرار گرفته است. معادلات جریان و حرارت به همراه مدل توربولانسی و مکانیزم شیمیایی مربوطه بوسیله یک الگوریتم ضمنی عددی و با استفاده از کد کامپیوتری که بدین منظور ساخته شده است حل شده اند. ابتدا مطالعه در جهت بررسی ساختار شعله و تغییرات آن نسبت به درجه حرارت هوای احتراق انجام گرفته است. سپس اثر درجه حرارت و غلظت O₂ بر انتشار NO_x از کوره مورد بررسی قرار گرفته است. نتایج نشان می دهد که با افزایش درجه حرارت هوای احتراق انتشار NO_x به شدت افزایش می یابد. افزایش راندمان احتراق و کاهش NO_x با افزایش درجه حرارت هوای ورودی به همراه کاهش همزمان غلظت O₂ امکان پذیر است. این موضوع ما را به سمت کوره های جدید با راندمان بالا و آلاینده گی کم هدایت می کند.

1. INTRODUCTION

NO and NO₂, abbreviated as NO_x, are emitted from combustion systems which use hydrocarbon fuel. It has been found that NO_x is a contributor of photochemical smog and ozone in the troposphere [1]. On the other hand, through a chain reaction it decreases the ozone layer in stratosphere, which may increase ultraviolet radiation on the earth [2]. Consequently, NO_x reduction has become a global demand that must be achieved by improving combustion science and technology.

As an effort in this regard, a technological method based on regenerative burning was proposed [3]. In this method, fresh airflow is diluted and heated up to 1200~1500 °K by circulation of exhaust gas through a heat exchanger. Experimental

reports [4] have shown many advantages for this technology. In addition to NO_x reduction, energy saving, high efficiency, and low noise are some of its advantages. However, although this technology has already applied for some industrial furnaces, its combustion mechanism has not been clarified yet. For instance, flame structure and NO_x formation mechanism are not well known yet [5].

In fact, the detailed process leading to NO_x formation even for laminar flame is not well understood yet [6,7]. There are many difficulties to be solved. The process for a simple hydrocarbon fuel like CH₄ involves about 100 intermediate species with more than 230 reactions. For any heavier hydrocarbon the number of intermediate species and corresponding reaction may reach about 2 or 3 times that of CH₄ fuel [8]. Furthermore,

when such process takes place in the turbulent flow the effect of the flow on the reaction and the interaction of turbulence-chemistry must be considered. Extinction caused by turbulence intensity has a vital effect on the combustion process. Moreover, for a non-premixed flame, turbulence has a significant effect on the mixing of reactant. Nevertheless, the recent increasing trend on research of NO_x emission is optimistic [8]. There have been numerous studies on NO_x formation mechanism in laminar flame [9,10], in turbulent flame [11], and in high-temperature air combustion [12,13].

For detailed study of the mechanism of NO_x emission in turbulent flow, a sophisticated method including fully detailed chemistry is needed. However, for a practical problem similar to what we are dealing with in this study, using a simplified chemical mechanism can qualitatively represent the fundamental characteristics of combustion phenomenon and NO_x formation.

As was mentioned above, the regenerative burner was experimentally and practically developed and was optimized for industrial furnaces. In order to study the NO_x emission of furnace in more details, numerical study was done on a simplified two-dimensional furnace model. The basic concept of the regenerative burner including of heating and diluting of fresh air by flue gas was implemented in this model. NO_x distribution in the furnace, and NO_x concentration at the flue gas were calculated for different conditions. Of special interest was the effect of temperature and O₂ concentration of preheated airflow on NO_x emission.

2. PHYSICAL MODEL

A simplified two-dimensional model of a heating furnace is shown in Figure 1. The model with a real scale is supplied with CH₄ fuel by two injectors of 5 mm, which are located up and lower of the airflow jet. The temperature of fuel and velocity of fuel and airflow are respectively, 300°K, 75 m/s, and 128 m/s. It is assumed that heating material flows on the lower wall, and the left, right and upper walls are at non-adiabatic condition. Based on the data obtained from an experimental furnace, temperature of left, right and upper walls were set at 1399 °K. Calculation

were done for four temperature of heating material, T_H = 300,600,1000,1324 °K, in which the last one was the maximum temperature obtained by the experimental furnace.

3. GOVERNING EQUATIONS AND NUMERICAL METHOD

The governing equations including two-dimensional, unsteady, compressible, Navier-Stocks equations for a multi-species gas undergoing chemical reaction can be written in the following forms:

$$\frac{\partial}{\partial t}(\rho v_i) + \frac{\partial}{\partial x_j}(\rho v_j v_i) = \frac{\partial}{\partial x_j} \left(\mu_e \frac{\partial v_i}{\partial x_j} \right) + S_i \quad (1)$$

$$S_u = -\frac{\partial p}{\partial x} + \frac{\partial}{\partial x} \left(\mu_e \frac{\partial u}{\partial x} \right) + \frac{\partial}{\partial y} \left(\mu_e \frac{\partial v}{\partial x} \right)$$

$$S_v = -\frac{\partial p}{\partial y} + \frac{\partial}{\partial y} \left(\mu_e \frac{\partial v}{\partial y} \right) + \frac{\partial}{\partial x} \left(\mu_e \frac{\partial u}{\partial y} \right)$$

Where

$$\mu_e = \mu_t + \mu$$

where u and v represent the velocities in x and y directions, respectively. The turbulent viscosity (μ_t) quantity can be obtained from the k-ε turbulence model[14]:

$$\frac{\partial}{\partial t}(\rho k) + \frac{\partial}{\partial x_j}(\rho v_j k) = \frac{\partial}{\partial x_j} \left(\frac{\mu_e}{\sigma_k} \frac{\partial k}{\partial x_j} \right) + S_k \quad (2)$$

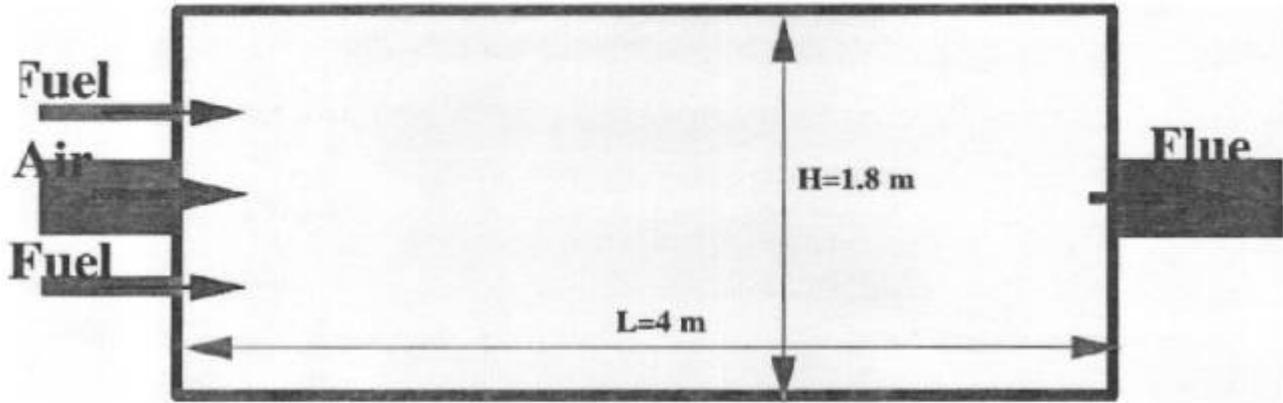
$$\frac{\partial}{\partial t}(\rho \epsilon) + \frac{\partial}{\partial x_j}(\rho v_j \epsilon) = \frac{\partial}{\partial x_j} \left(\frac{\mu_e}{\sigma_\epsilon} \frac{\partial \epsilon}{\partial x_j} \right) + S_\epsilon \quad (3)$$

Where

$$S_k = G_k - \rho \epsilon$$

$$S_\epsilon = \frac{\epsilon}{k} (c_1 G_k - c_2 \rho \epsilon)$$

$$G_k = \mu_t \left[2 \left(\frac{\partial u}{\partial x} \right)^2 + 2 \left(\frac{\partial v}{\partial y} \right)^2 + \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right)^2 \right]$$



$$\begin{array}{ll}
 D_{\text{air}}=0.39 \text{ m} & D_{\text{fuel}}=0.005 \text{ m} \\
 V_{\text{air}}=128 \text{ m/s} & V_{\text{fuel}}=75 \text{ m/s} \\
 & T_{\text{fuel}}=300 \text{ K}
 \end{array}$$

Figure 1. Schematic of physical model.

$$\mu_t = c_\mu \rho k^2 / \varepsilon$$

$$C_\mu = 0.09, \quad C_1 = 1.44, \quad C_2 = 1.92, \quad \sigma_k = 1.0, \\ \sigma_\varepsilon = 1.33$$

Diffusion equation can be written as:

$$\frac{\partial}{\partial t}(\rho m_i) + \frac{\partial}{\partial x_j}(\rho v_j m_i) = \frac{\partial}{\partial x_j} \left(\rho D \frac{\partial m_i}{\partial x_j} \right) - \omega_i \quad (4)$$

where m_i is the mass fraction of the i th species. Energy equation is given as the following form:

$$\rho c_p \frac{DT}{Dt} = \nabla \cdot (\lambda \nabla T) - \sum_{i=1}^N \omega_i h_i - \sum_{i=1}^N (C_{p_i} \nabla T \cdot \rho m_i v_i) \quad (5)$$

An overall one-step reaction mechanism was used for CH₄ oxidation [18].



$$\omega = K_1 [\text{CH}_4]^2 [\text{O}_2]^3$$

$$K_1 = 6.7 \times 10^{12} \exp\left(-\frac{48.4 \text{ kcal/mol}}{RT}\right)$$

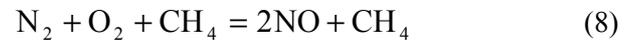
Furthermore, for thermal NO the mechanism suggested by Beltagui [19], and for prompt NO the mechanism recommended by Missaghi [20] were used;

Thermal NO:



$$\frac{d[\text{NO}]}{dt} = 1.44 \times 10^{17} T^{0.5} [\text{N}_2] [\text{O}_2]^{0.5} \exp\left(-\frac{69460}{T}\right)$$

Prompt NO:



$$\frac{d[\text{NO}]}{dt} = f \cdot T^\beta \cdot A \cdot [\text{N}_2] [\text{O}_2]^{0.5} [\text{CH}_4]^{0.5} \exp\left(-\frac{E}{RT}\right)$$

$$E = 303 \text{ kJ/mol} \quad A = 6.4 \cdot 10^6 (RT/P)^{a+1}$$

$$f = 4.75 + C_1 n + C_2 \phi + C_3 \phi^2 - C_4 \phi^3$$

where n is the number of carbon atoms per molecule,

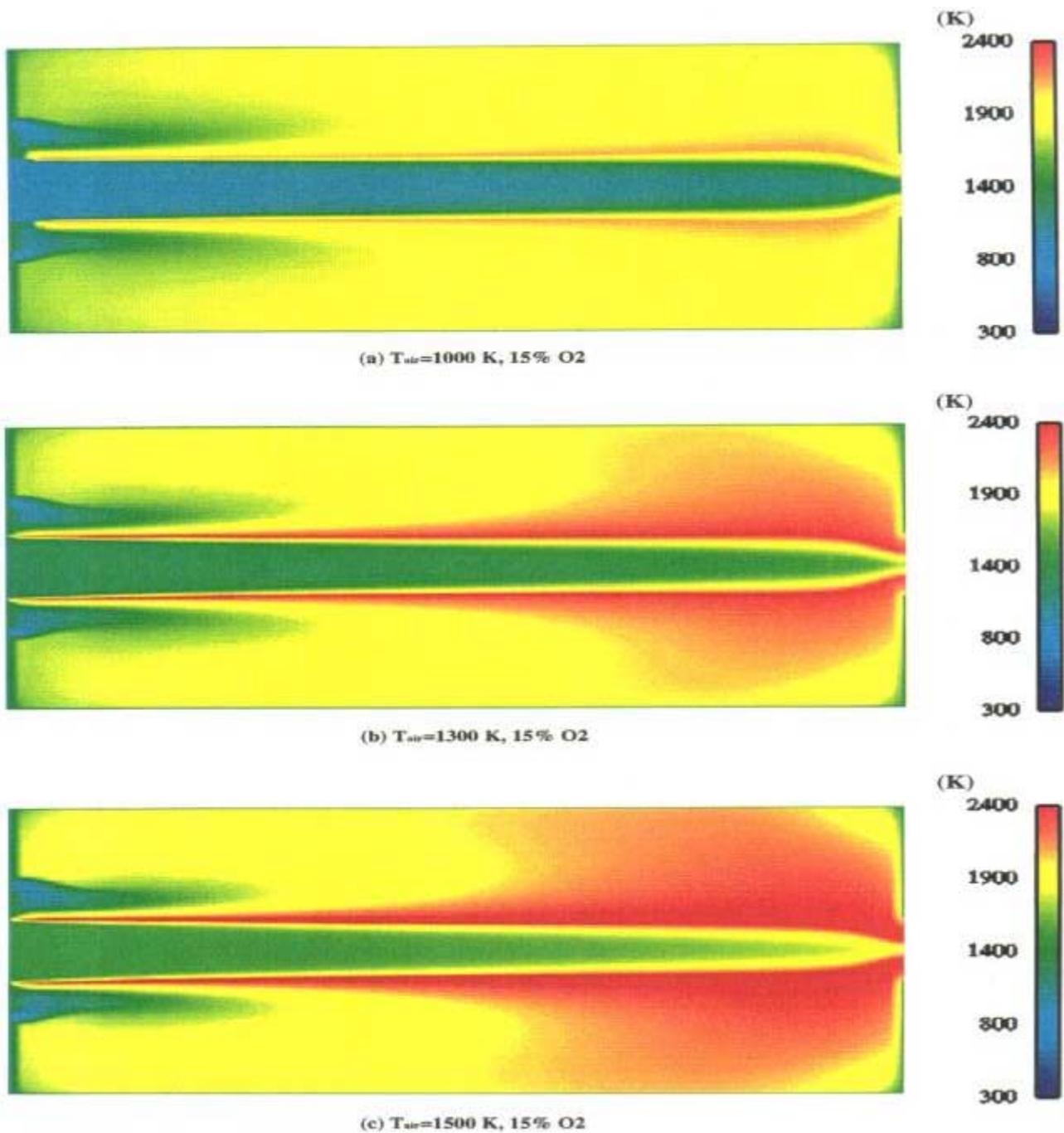


Figure 2. Temperature distribution in furnace in different airflow temperature.

and ϕ is the equivalence ratio, C_i is constant.

For solving the governing equations the well-known SIMPLE scheme of Patankar [15] was used. Specific heat, C_p , for each species at a constant pressure was computed by a fourth-order polynomial

fitted to the data tabulated in the JANAF tables [16]. The molecular viscosity, thermal conductivity and diffusion coefficient were determined by the methods, which are recited in Reference 17.

An H-type grid system with 102×158 grid points

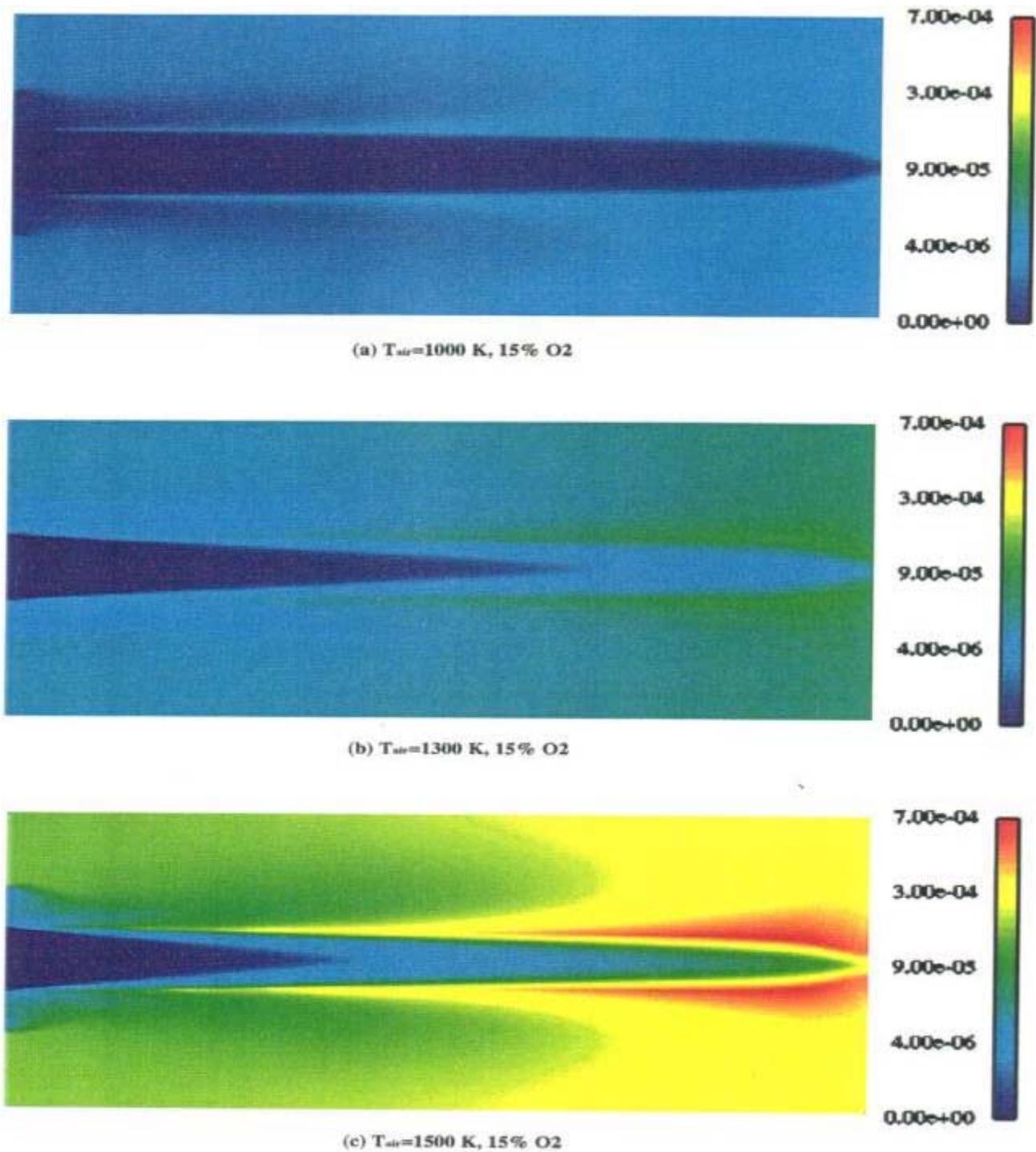


Figure 3. Distribution of NOx mass fraction in furnace in different airflow temperature.

were used for the entire of the computational domain. 102×6 grid points just were used for fuel nozzle region to ensure high resolution at this

region. Also, 102×30 grid points were inserted in the airflow jet region. A high-resolution grid mesh was used at the interface of jet flows and near the

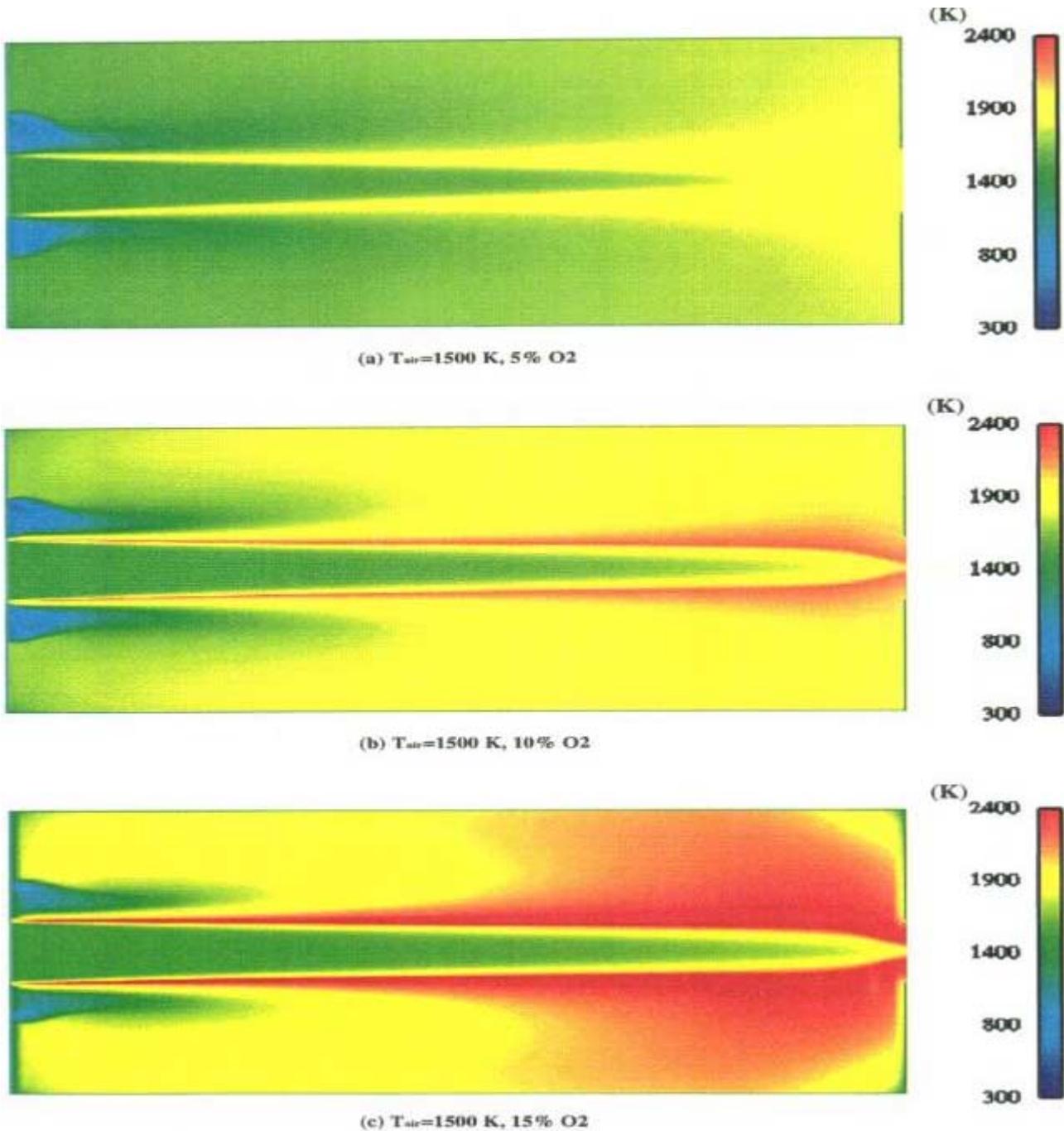


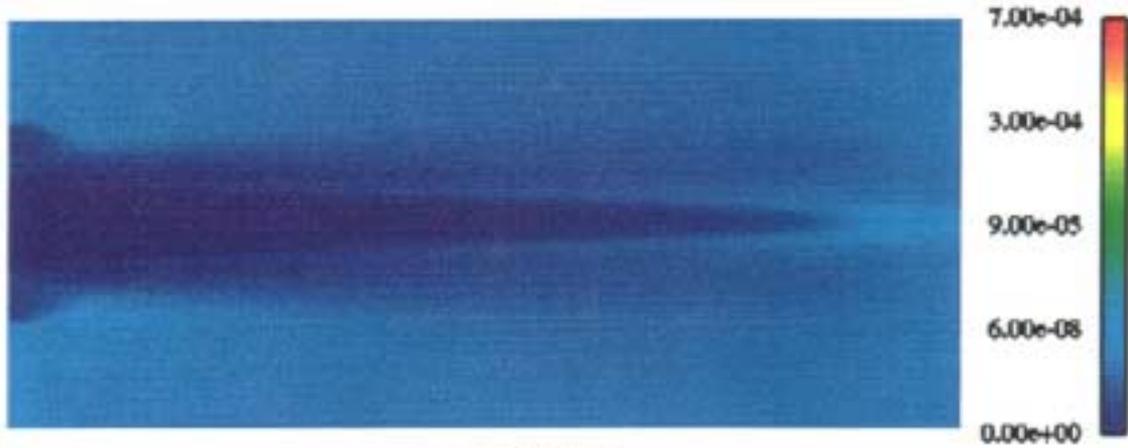
Figure 4. Temperature distribution in furnace in different O₂ concentration.

walls where the gradient is sharp.

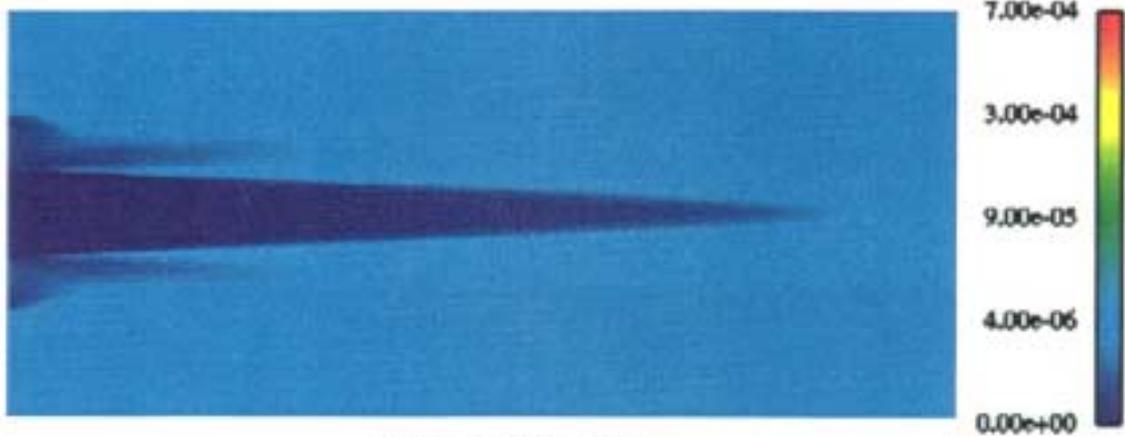
4. RESULTS AND DISCUSSION

Three kinds of calculation were done in this study.

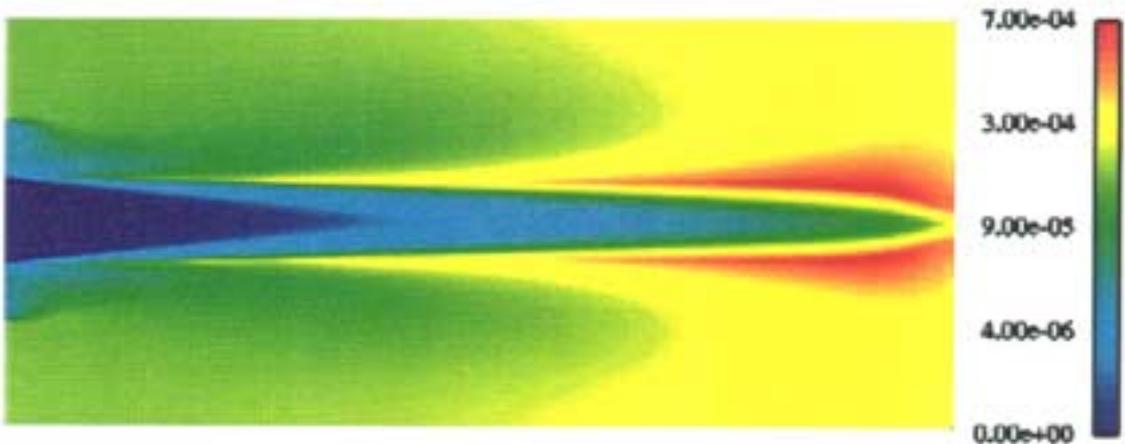
First keeping O₂ mass fraction constant the effect of temperature of preheated airflow was investigated. Second, keeping temperature of preheated airflow at $T_{air} = 1500 \text{ }^\circ\text{K}$, calculation was conducted to study the influence of O₂ reduction on NO_x emission. And finally, considering that flue gas circulation



(a) $T_{in}=1500$ K, 5% O₂



(b) $T_{in}=1500$ K, 10% O₂



(c) $T_{in}=1500$ K, 15% O₂

Figure 5. Distribution of NO_x mass fraction in furnace in different O₂ concentration.

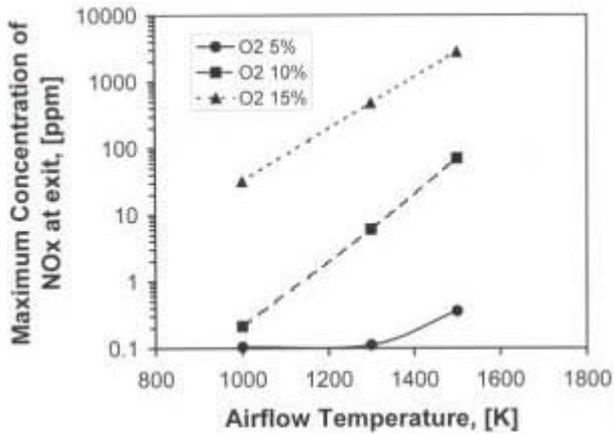


Figure 6. Variation of maximum concentration of NO_x at the exit of furnace against temperature of preheated airflow for different O₂ concentration.

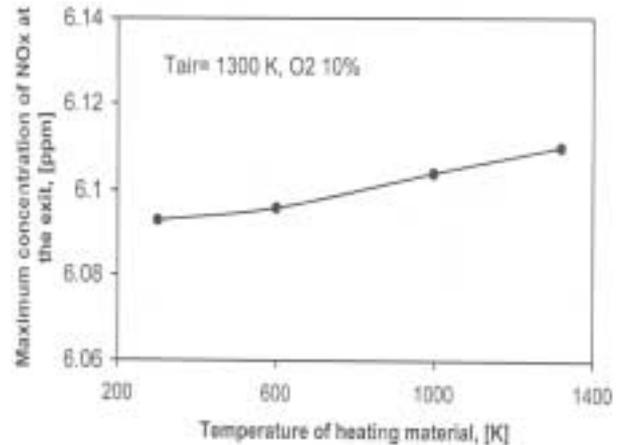


Figure 8. Variation of maximum concentration of NO_x at the exit of furnace against temperature of heating material.

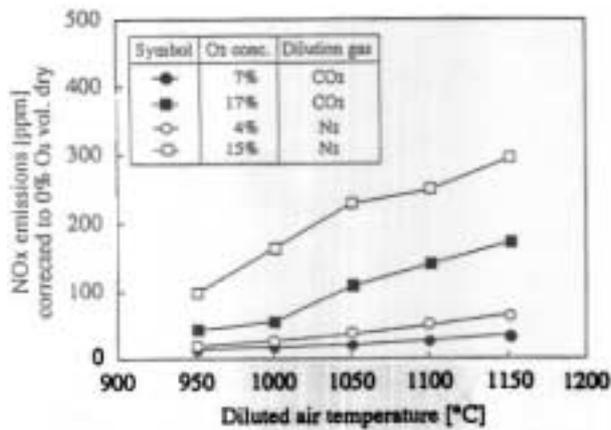


Figure 7. Experimental results by Hasegawa et al. [4].

is done at high temperature, preheated airflow may enter in the furnace at equilibrium or semi-equilibrium condition. At such condition there should be a considerable amount of NO_x in the preheated airflow. Calculation was done to investigate the effect of such pre-produced NO_x on NO_x emission.

In Figures 2 and 3 distribution of temperature and NO_x mass fraction are shown, respectively. The fuel temperature was kept constant at 300 °K, while the temperature of preheated airflow is changed from 1000 to 1500 °K. As it can be seen in Figure 2, with increasing of T_{air}, the high temperature zone is expanded in the furnace and

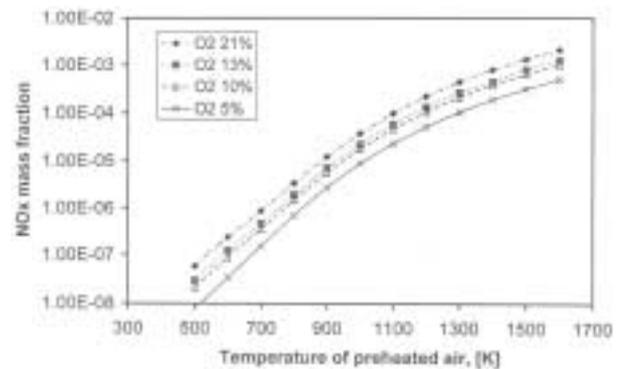


Figure 9. NO_x mass fraction against temperature of preheated air at equilibrium condition.

flame temperature significantly increases. As a result, NO_x emission drastically rises, so that, its peak amount at the furnace exit for T_{air} = 1500 °K becomes more than 85 times of it for T_{air} = 1000 °K. However, the combustion efficiency for these two cases stands at 90.5% and 81.3%, respectively. Here combustion efficiency was calculated as the ratio of fuel consumption to inlet fuel. As Figure 2 shows even for preheated airflow of T_{air} = 1000 °K, flame temperature in about 60% area of the furnace is more than 1800 °K. At such high temperature condition, formation of NO_x is significantly dominated by thermal NO. Figure 3 clearly shows that, the more the temperature increases the more the NO_x mass fraction raises in the furnace. However, at relatively low flame temperature

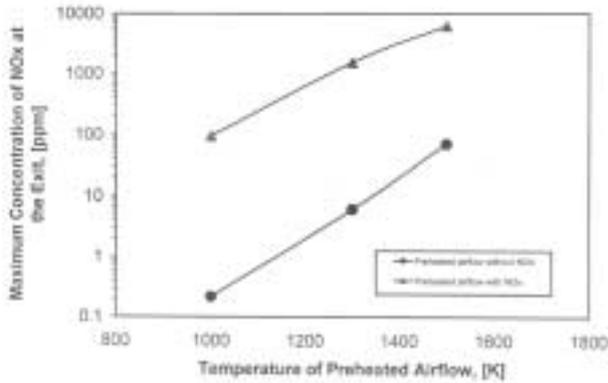


Figure 10. Variation of maximum concentration of NO_x at the exit of furnace against temperature of preheated airflow for preheated airflow with and without NO_x .

area in the furnace (about 1000 °K) formation of thermal NO is negligible because of its high activation energy. At such condition, prompt NO is produced, particularly, at fuel-rich condition.

Calculation was conducted to investigate the effect of O_2 concentration of preheated airflow on emission of NO_x in the furnace. Figures 4 and 5 show the temperature distribution and NO_x mass fraction, respectively. Airflow was diluted by CO_2 to reduce O_2 concentration, and temperature of preheated airflow was kept at 1500 °K, while fuel condition was the same as before. As it can be seen in Figure 4, by reducing O_2 concentration of preheated airflow the high temperature zone becomes smaller and the flame temperature decreases considerably. For the typical cases shown in Figure 4, a 50% reduction of O_2 causes about 20% decrease in flame temperature. O_2 deficiency is one of the responsible factors for flame temperature reduction. However, the injection of dilution gas such as CO_2 whose additional heat capacity lowers the peak temperature is another important factor. As a result of flame temperature reduction, NO_x concentration reduces sharply as shown in Figure 5.

Variation of maximum concentration of NO_x at the exit of furnace against temperature of preheated airflow at different O_2 mass fraction is shown in Figure 6. It can be clearly seen that by reduction of O_2 mass fraction, NO_x concentration sharply drops at any temperature of preheated airflow. Furthermore, it can be seen that when

temperature of preheated airflow increases from 1000 °K to 1500 °K, NO_x concentration increases 3.5 times for O_2 5%, but for O_2 10%, NO_x concentration increases 330 times. In fact, such behavior is a primary effect of high heat capacity of CO_2 . Experimental result [4] clarified that if N_2 is used as dilution gas instead of CO_2 , NO_x emission increases more than 2 times. Considering that the heat capacity of CO_2 is much larger than N_2 one can conclude that if a dilution gas with heat capacity more than CO_2 is used, NO_x emission even becomes smaller.

The other fact that may reveal the mechanism of NO_x reduction in Figure 6 is the effect of O_2 reduction on combustion regime. Reducing O_2 concentration in the furnace shifts the combustion regime to the fuel-rich condition. And we know that NO_x formation at the fuel-rich is much smaller than stoichiometry flame, particularly at lower temperature.

In a heating furnace, heating material with initial temperature of 300 °K are heating up to about 1200~1300 °K. Of our interest is to find the effect of such temperature variation on NO_x emission. For a typical preheated airflow of 1300 °K and O_2 10%, calculation was done at 4 different temperatures of heating material, T_H , while keeping other walls temperature constant at 1399 °K. The results of maximum concentration of NO_x at the exit of furnace are shown in Figure 7 when temperature of heating material rises to 1300 °K from its initial temperature of 300 °K, only there is about 0.3% increase in NO_x concentration.

As was mentioned before, preheated airflow may enter in the furnace at equilibrium condition. At such condition there should be a considerable amount of NO_x in preheated airflow. Calculation was done to investigate the effect of such phenomena on NO_x emission from the furnace. First, NO_x formation in high temperature airflow at equilibrium condition was calculated [21]. The results of this calculation are shown in Figure 8. The NO_x mass fraction gradually increases by increasing of temperature of airflow, and as it is expected, reduction of O_2 concentration results in lower NO_x formation; Figure 8. By injection preheated airflow at equilibrium condition with NO_x mass fraction given in Figure 8, calculation was done for O_2 10% at three temperature of

preheated airflow; 1000, 1300 and 1500 °K. The calculated results are compared with the results of injection of preheated airflow without NO_x in Figure 9. Clearly, it shows a drastic effect on NO_x emission from the furnace. However, further investigation showed that the inlet NO_x, which is produced at the equilibrium condition, directly causes the increasing of NO_x in the furnace. Therefore, the flame temperature in the furnace not only doesn't increase but in fact slightly decreases.

5. CONCLUSION

NO_x emission from a furnace utilizing regenerative burner technology was numerically investigated. Influences of inlet airflow temperature and reduction of O₂ concentration of preheated airflow on NO_x formation were studied. Calculations were also done to consider the effects of heating material temperature and pre-production of NO_x in the heat exchanger on NO_x emission from the furnace.

Numerical results showed a drastic increase in NO_x emission as temperature or O₂ concentration of preheated airflow increases. A furnace with high efficiency and low NO_x emission can be achieved when the inlet airflow is preheated up to a high temperature, about 1500 °K, and O₂ reduced to about 5%.

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