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Experimental and Numerical Study on the Effect of Dilution in Premixed CH₄-Air Flame

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Abstract

Dilution is considered as a method, which reduces the temperature of furnace and decreases the formation of thermal NO. All of the NO_x controlling methods are based on the control of temperature or limiting the concentration of O₂. In this study, the numerical and experimental effects of N₂ and CO₂ diluents on a premixed methane–air flame in a furnace are investigated. This experimental study is achieved by use of a cylindrical furnace for an equivalence ratio from 0.6 to 1.4 and different dilution ratios. The simulation is carried out by using a premix code of the CHEMKIN II software. The GRI-Mech 3.0 detailed reaction mechanism is employed for the combustion of methane. Numerical and experimental results show that the maximum temperature of a flame and the rate of NO_x production are reduced by the increase of the dilution ratio. The results also demonstrate the fact that a CO₂ diluent is more effective than an N₂ due to its high thermal capacity that reduces the NO_x production rate more than the N₂ can.

Keywords: Dilution, NO_x, Experimental, Numerical, Premixed.

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1. Introduction

The combustion of fossil fuels leads to significant pollution and is probably the major contributing factor in global warming. Scientists and engineers are working to develop alternative energy sources in order to reduce society's reliance on oil and to minimize pollutants and greenhouse gas emissions. Fossil fuels are nonrenewable and a major source of pollutants, including CO₂, NO_x, UHC and soot (Briones et al. [2]). When fossil fuels (such as natural gas and oil) and their derivative compounds burn with air, oxidation products, like CO, CO₂ and H₂O, are formed. In addition, nitrogen molecules in the air react with some oxygen molecules and form nitrogen oxides (Schefer et al. [29]). Nitrogen oxides include nitric oxide (NO), nitrogen dioxide (NO_2) and nitrous oxide (N_2O) , of which NO and NO₂ are known as NO_x (Hill & Smoot [11]). NO is a toxic gas which is formed during combustion at high temperature zones in a combustion chamber (i.e. automobile engines, power plants and furnaces); while entering the atmosphere, NO changes into NO_2 . Generally, three mechanisms for the production of NO in combustion are considered: thermal NO, fuel NO and prompt NO. NO that is produced from nitrogen and oxygen molecules in the air are called thermal NO and that which is produced from nitrogen molecules in fuel is called fuel NO. The NO concentration near the flame zone of hydrocarbon fuels is less dependent on temperature and radicals, like O and OH, which help to form NO. Thus, in spite of the control by flame temperature and oxygen concentration, NO forms and is called immediate NO (prompt NO) (Fenimore). The thermal mechanism, which is also called the Zeldovich [33] mechanism, is the main route in the methanehydrogen-air premixed flame since a suitable condition for the production of prompt NO and fuel NO is not available (Warnats et al. [31]). The thermal NO consists of the following reactions:

Vol. 4(12), Jul, 2014, pp. 1155-1174, ISSN: 2305-0543 Available online at: http://www.aeuso.org © Austrian E-Journals of Universal Scientific Organization $O + N_2 \rightarrow NO + N$ $N + O_2 \rightarrow NO + N$ (1) $N + OH \rightarrow NO + H$

The first reaction has a high activation energy because of the strong triple bond of N_2 . Therefore the reaction takes place at a high temperature; otherwise, it will limit the formation of thermal NO. Equation (2) is used for the concentration of NO (Warnats *et al.* [31]).

$$[NO] = K_1 \exp\left(\frac{-K_2}{T}\right) [N_2] [O_2]^{\frac{1}{2}} . t$$
(2)

where K_1 and K_2 are the Reaction kinetics constants and t is time. T is temperature and [NO] is concentration. The thermal mechanism of NO is highly sensitive to temperature as an exponential function. At temperatures higher than 1700 K, the formation of NO will double per each 40 degrees increase so the techniques which reduce the flame temperature are effective for the reduction of NO (Lazic *et al.* [17]).

Dilution, as a way of reducing of NO_x is considered in many experiments since it shifts the combustion to the lean condition and reduces the formation of nitrogen oxides (Rortveit *et al.* [26] and Ghoniem *et al.* [9]). Adding a diluent with a high heat capacity increases the total heat capacity of the fuel–air–diluent mixture and, as a result, the mixture absorbs more heat during the combustion and reduces the temperature of the combustion chamber. This leads to the reduction of NO_x emission production which is related to the temperature decrease (Kim *et al.* [15]). Until now, there have been various types of diluents which have been studied in the field of combustion, such as CO₂, N₂, H₂ and H₂O, along with their effects on the combustion parameters. Li & Williams [20] conducted an experimental and numerical study on the effects of air stream dilution using various agents (N₂, H₂O, CO₂ and Ar) on NO_x emission from CH₄–air counter flow flames. On a mass basis, the effectiveness of

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various diluents in reducing NO_x emission was found to rank in this order: H₂O>CO₂>N₂>Ar. The greater effectiveness of H₂O was due to its higher heat capacity that lowers the flame temperature and thereby thermal NO, in addition to its ability to decrease the concentration of CH radicals that reduce prompt NO. Lee et al. [18] studied the effects of including CO₂ in an oxidizer stream on the flame structure and on NO_x formation in a CH₄-air counter flow non-premixed flame. Liu *et al.* [21] studied the chemical effects of CO₂ as an additive on soot and NO_x formations in an ethylene non-premixed flame. All of these studies focused on the molecular transport and chemical kinetic characteristics of CO_2 dilution, which was thought to be mainly explained through chemical and thermal effects. In addition to these effects, however, the hydrodynamic effects due to CO₂ dilution can change many fundamental phenomena, such as entrainment and mixing, local extinction due to flame stretching, diffusion and differential diffusion and vorticity distribution in a reacting flow field (Chen et al. [3]). Zhao et al. [34] numerically examined the effects of the steam addition on NO_x formation in methane–air diffusion flames. A 17% reduction in NO_x emission index with a 5% mass addition was observed, while the same flame temperature was maintained. Rortveit et al. [27] reported experimental and numerical results on the effects of various diluents in the fuel stream on NO_x emissions from H₂-air non-premixed and partially premixed flames. On a mole basis, CO₂ was found to be a more effective diluent compared to N₂ in reducing both the flame temperature and the NO_x concentration in these flames. Park *et al.* [23] also investigated the effect of steam addition on NO formation in H2-O2-N2 diffusion flames. Salvador et al. [28] studied the effect of dilution using N_2 on NO_x emission in a furnace with natural gas as fuel. Their results showed N_2 dilution reduces NO_x emission by more than 60%. Cho et al. [4] numerically investigated the effect of N_2 and CO_2 diluents on NO_x reduction. It was concluded that diluting with CO_2 is more effective in NO reduction when compared to N₂, because of the large temperature drop due to the greater

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specific heat of CO₂. Prathap *et al.* [24] studied the effects of nitrogen dilution on the laminar burning velocity and flame stability of syngas fuel. Their results indicated that nitrogen dilution decreased the laminar burning velocity due to a reduction in heat release and increased the heat capacity of the unburned gas mixture and, hence, the flame temperature. Giles *et al.* [10] numerically investigated the effect of dilution on a non-premixed flame using H₂O, CO₂ and N₂ as a diluent. It was reported that H₂O and CO₂, due to their higher heat capacities, are more effective than N₂ for reducing NO_x emission and the temperature of the combustion chamber. Kobayashi *et al.* [16] explored the dilution effects of superheated water vapor on turbulent premixed flames at a high pressure and temperature. These studies revealed that water vapor plays an important role in reducing of NO_x emission. Other findings reported that hydrogen enrichment alone leads to higher burning velocities (Coppens *et al.* [5]), while others indicated that dilution by nitrogen leads to a decrease in the burning velocity (Dyakov *et al.* [7]) and to a lower decrease in NO_x emissions (Turns *et al.* [30], Dyakov *et al.* [8]).

2. Experimental

In order to investigate the effect of fuel dilution on the formation of pollutants NO_x , a furnace was designed and constructed. The main body of the furnace is a hollow tube of steel AISI 316 with a diameter of 105 mm and a length of 1000 mm to with stand high temperatures. The air needed for combustion is supplied by a fan that is capable to change the mass flow rate of air. Fuel and diluents pass through a venturi in order to achieve a better mixture and after passing through a 35 mm wide tube, enter the furnace. This also prevents a possible flash back. The furnace is insulated by a layer of Indian cotton 1260 with a 15 mm thickness. In order to measure the temperature and observe the flame, 15 holes with a diameter of 20 mm and 70 mm apart from each other, were placed above the furnace body. For better

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mixing of fuel and diluent, a collector is used. The flow rate of the fuel and diluents is measured before entrance to the collector and then they are allowed to enter the collector. After entrance to the collector, the rate is measured again. The collector was optimally designed to accommodate a short circuit input and an entrance by the fuel and diluents at an angle under 45 degrees. Figure 1 shows the schematic of the designed furnace and its accessories.



Figure 1. Schematic of experimental setup

To reduce pressure inside the CO_2 cylinder, (70 bar), a regulator is used which reduces the pressure to 2.5 bar (optimal pressure). Also, to reduce pressure inside the N_2 cylinder and adjust it from 140 bar to the desirable level, MORRIS barometers were used, there are capable of tolerating 200 bar of input pressure and can reduce pressure to 1 bar. A gas analyzer, the TESTO 350 XL model with an accuracy of 0.05 ppm, was chosen to measure

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the NO_x of the exhaust gases. A flow meter (Lutron YK-2005AM), with an accuracy of 0.01 to 0.05 m/s, was employed to measure the mass flow rates of the air and exhaust gas, in addition to the mass flow rate of the fuel and diluent. Two SWPF-06A rotameter, with an accuracy of 0.02 lit/min, were utilized. Because of their accuracy, type R thermocouples (platinum-rhodium alloy containing 13% rhodium for one conductor and pure platinum for the other conductor), measured the absolute temperature within a tolerance of \pm 10°C and the relative temperatures within \pm 1°C.

3. Experimental Errors

Regardless of the accuracy of experiments, errors may occur because of various reasons when collecting empirical data. Some errors types are random and some are due to pitfalls during the test. Obvious errors resulting from incorrect data are immediately identifiable and have been excluded. However, there are data that are suspicious and not easily identified. Therefore, to identify these errors, an uncertainty analysis of empirical data obtained via statistical method, was used. The amount of uncertainty or possible error, depending on test conditions, may change considerably. Fixed and random errors are the errors that may cause uncertainty in experimental measurements. Often, fixed errors can be corrected by the calibration of equipment, but random errors must be identified through statistical analysis. In this research, for each point, first four types of laboratory data are collected and then the arithmetic mean of data is calculated. The amount of potential error or uncertainty of data is calculated from the following formula:

$$\sigma_m = \frac{\sigma}{n^{0.5}} \tag{3}$$

In this formula, σ_m is the standard deviation of the mean value, σ is the standard deviation measurement and *n* is the set of numbers of measurement at any point for

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all the experimental results. The effect of dilution with CO_2 and N_2 , is 4. The amount of the experimental data uncertainty percentage, (X), is calculated from the following equation:

$$X\% = \frac{\sigma_m}{\overline{x}} \times 100,\tag{4}$$

where \overline{x} is the arithmetic mean value of experimental data. Figure 2 shows the average uncertainty of experimental data in the furnace output.



Figure 2. Average uncertainty values for NO_x output and the temperature of furnace methane-diluted air in the range of equivalence ratios and dilution ratios: CO_2 ; N_2

4. Computational Method

The effect of dilution on a premixed methane–air flame is simulated using the commercially available CHEMKIN II package (Kee *et al.* [12]), more specifically, the Premix code (Kee *et al.* [13, 14]). The detailed reaction mechanism employed GRI-Mech 3.0 (Bowman *et al.* [1]) consisting of 325 elementary chemical reactions and 53 species describing methane oxidation chemistry. Multi component diffusion and thermal diffusion options were taken into account. The adaptive mesh parameters were GRAD=0.05 and CURV=0.5. The relative and absolute error criteria were RTOL=1×10-5 and ATOL=1×10-5.

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9, respectively; the total number of grid points was typically 350–400. The steady state mass, momentum, energy, and species conservation equations were solved along the stagnation streamline in a finite domain (Ren *et al.* [25]).

5. Results

The numerical and experimental study was conducted over a wide range of dilution and equivalence ratios. The dilution ratio (β) is computed from:

$$\beta = \frac{n_{\text{Diluent}}}{n_{\text{Fuel}}},\tag{5}$$

where $n_{Diluent}$ and n_{Fuel} are the number of diluent moles and fuel moles. Figure 3 shows the effect of dilution with CO₂ and N₂ diluents at a maximum flame and furnace temperature in various equivalence ratios.



Figure 3. Effect of CO₂ and N₂ diluents on temperature; (experimental and numerical)

As it can be seen in Figure 3, the addition of diluents decreased the temperature of the furnace. Also, these figures indicate a decrease of the maximum temperature of the flame with the addition of diluents. The maximum flame temperature (numerical results (lines)) reached maximum values at $\varphi = 1.0$, because of the presence of a sufficient amount of O₂.

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The temperature of the furnace's maximum values (experimental results (points)) occurs at about $\varphi = 0.95$, because of the presence of the diluent's and hydrodynamic effects leading to a shift in the combustion to the lean condition. Because of the diluents presence in the fuel–air mixture, the thermal capacity of the mixture increased and consequently the flame temperature and chamber temperature decreased. The breaking process of the N₂ bond is endothermic, thus reducing the flame temperature and the formation of NO_x. Figure 4 shows the simulation results of the effect of CO₂ and N₂ diluents in the premixed methane– air flame at $\varphi = 1.0$.



Figure 4. Effect of CO₂ and N₂ diluents on flame temperature at $\varphi = 1.0$ (numerical)

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Figure 5. Effect of φ on NO_x emission and a comparison with the other researchers The decrease of the flame temperature with CO₂ diluent is more remarkable than that of

 N_2 because of CO₂'s higher thermal capacity. The thermal capacities of CO₂ and N_2 are 2.9 kJ/kg.K and 1.042 kJ/kg.K respectively. Figure 5 illustrates the numerical and experimental results of the effect of the equivalence ratio on NO_x formation in a premixed methane–air flame without dilution. The results also compared with other researchers.

From the figure it can be seen, NO_x formation increases with that of the equivalence ratios, from 0.6 to its peak at 0.95. After that, NO_x formation is reduced with the increase of the equivalence ratios in a rich mixture. This conforms to the temperature trend. Temperature is the main parameter in NO_x formation because of its exponential trend (Lazic *et al.* [17]). The concentration of O_2 and N_2 in a lean mixture may is reduced but the combustion moves toward a complete one in which the flame temperature reaches its peak.

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The lack of O_2 in a lean mixture diminishes the oxidization process of N_2 . As a result, a rise in temperature will cause the NO_x to increase.

Figure 6 shows the experimental results of NO_x concentration over a wide range of equivalence ratios for various dilution ratios in a premixed methane—air flame. The presence of a diluent mixture increased the thermal capacity and reduced the oxygen mole fraction at the entrance, which will lower the flame temperature and NO_x emissions. All the factors that diminish the maximum flame temperature and eventually the temperature of furnace are considered effective in reducing NO_x . Also, with reduction of the oxygen mole fraction, the partial pressure will drop, which will cause the fundamental reaction kinetics and NO_x production rate to decrease.



Figure 6. Effect of CO2 and N2 diluents on NOx emission (experimental)

Experimental results show that in $\varphi < 1.0$ (lean mixture), N₂ and CO₂ dilution lead to a decrease of the mass fraction of fuel in the mixture, which, in turn, causes flame extinction. This flame extinction also occurs in $\varphi > 1.0$ (rich mixture) due to the reduction of air. Table 1 shows the extinction limitation of the flame in different equivalence ratios.

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Table 1. Flame extinction limits in various equivalence ratios ($0.0 < \beta_{co.} < 1.0$ and

equivalence ratio (φ)	CO ₂ (0.0<β<1.0)	N ₂ (0.0<β<2.0)
0.6	0.4	0.8
0.7	0.6	1.6
0.8	0.75	-
0.9	-	-
1.0	-	-
1.1	-	-
1.2	0.75	1.7
1.3	0.7	1.55
1.4	0.4	1.3

$$0.0 < \beta_{_{N_2}} < 2.0$$
)

Figure 7 shows the numerical and experimental results of the effects of CO_2 and N_2 diluents on NO_x emissions for different equivalence ratios.

As it can be seen from these figures, with the increase of the dilution ratio, NO_x emission decreases due to the increased heat capacity of fuel, air and the diluent mixture. In addition, the experimental results at the equivalence ratios of 0.6, 0.7, 0.8, 1.2, 1.3, in which extinction occurs, are not available. Table 2 shows the percentage of NO_x reduction up to extinction (if it occurs) at different equivalence ratios.

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13 3.1 Experimental (CO2) Experimental (CO2) 12 2.9 -D-Numerical (CO2) −□− Numerical (CO2) 11 . 2.7 ▲ Experimental (N2) ▲ Experimental (N2) 10 2.5 ∆–Numerical (N2) -∆-- Numerical (N2) 2.5 (uidd) XON 1.7 N 1.5 1.3 9 $\phi = 0.7$ D=0.6 9 8 7 6 5 4 MOX (mdd) 3 1.1 2 0.9 0.7 1 0 0.5 0.2 0.8 1.2 1.6 1.8 2 2.2 0 0.2 0.4 0.6 0.8 1.2 1.6 2 2.2 0 0.4 0.6 1 1.4 1 1.4 1.8 β ß 70 50 Experimental (CO2) Experimental (CO2) 45 -D-Numerical (CO2) 60 $\varphi = 0.8$ - Numerical (CO2) $\varphi = 0.9$ ▲ Experimental (N2) 40 ▲ Experimental (N2) Numerical (N2) - Numerical (N2) 50 35 (uddd) xON 20 (udd) xON 15 10 10 5 0 0 0.2 0.6 0.8 1.2 1.4 1.8 2 2.2 0 0.4 1 1.6 0 0.2 0.4 0.6 0.8 1 1.2 1.4 1.6 1.8 2 2.2 ß ß 70 110 Experimental (CO2) Experimental (CO2) 100 60 $\varphi = 1.1$ -D-Numerical (CO2) $\varphi = 1.0$ ▲ Experimental (N2) ▲ Experimental (N2) 90 50 ∆—Numerical (N2) ≜–Numerical (N2) (uidd) xON 20 80 (udd) xON 40 10 30 20 0 0 0.2 0.4 0.6 0.8 1 1.2 1.4 1.6 1.8 2 2.2 0 0.2 0.4 0.6 0.8 1 1.2 1.4 1.6 1.8 2 2.2 β ß

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Figure 7. Effect of CO₂ and N₂ diluents on NO_x emission (experimental & numerical)

	NO _{<i>x</i>} reduction (%)				
φ	CO ₂		N_2		
	Experimental	Numerical	Experimental	Numerical	
0.6	40^{*}	26	30*	44	
0.7	61*	62	67*	48	
0.8	74 [*]	89	46	57	
0.9	85	86	58	61	
1.0	71	57	33	45	
1.1	88	84	62	74	
1.2	63*	84	55*	76	
1.3	62*	63	68^*	68	

Table 2. Percentage of NO_x reduction for CO₂ and N₂ diluents

*Extinction occurs

Conclusion

In this study the effects of dilution on NO_x formation are investigated numerically and experimentally. Results are as follows:

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- With fuel dilution, the temperature of a furnace decreases due to the increased thermal capacity of the mixture and the reduction of the oxygen concentration.
- The maximum concentration of temperature and NO_x in the premixed flame without dilution occurs at the equivalence ratio of 0.95.
- With an increase of the dilution ratio, the temperature of the furnace and, consequently, NO_x formation is reduced.

The decrease of the flame temperature and NO_x with the CO_2 diluent is more remarkable than that of N_2 because of CO_2 's higher thermal capacity.

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