



The Effect of the Flue Gas Recirculation on NO_x Formation in a Premixed Methane/Air Flam

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Abstract

The reaction of thermal NO_x is highly sensitive to temperature and if the technique can reduce the flame temperature it would be effective to reduce NO_x formation. Flue gas recirculation can reduce the flame temperature and as a result decrease the rate of NO_x formation. In the present study the effect of FGR on NO_x emission in the premixed methane-air mixture is investigated numerically and experimentally. The experiments are carried out in an axial symmetry cylindrical combustion chamber for a range of equivalence ratio and different percent of FGR. The numerical simulations are conducted simultaneously, using Premix code of Chemkin package. The GRI-Mech3.0 mechanism is used for the combustion of methane. The results reveal that as the percentage of flue gases is increased, the flame temperature and NO_x are decreased. This is because of the presence of species with high thermal capacity in flue gases. The experimental results show that the

NO_x emission is decreased by 66 percent by addition of 30 percent of the flue gases. The simulation results are in good agreement with the experimental results.

Keywords: Flue Gas Recirculation, NO_x, Methane/Air Flam

1. Introduction

NO₂ and N₂O have the most damaging impact on the atmosphere. Gas-fired furnaces and boilers produce mainly NO, but in contact with the air, the NO gradually oxidizes into NO₂ [1]. There are three mechanisms for the formation of NO_x in combustion processes: thermal NO_x, prompt NO_x and fuel NO_x. Thermal NO_x is formed from molecular nitrogen at very high temperature. Prompt NO_x is formed in fuel-rich region, where hydrocarbon radicals react with molecular nitrogen to form NO_x at high temperature. However, the possibility of prompt NO_x is usually negligible, because industrial furnaces mostly run under oxygen-rich conditions. Fuel NO_x is formed from nitrogen atoms bound in the fuel [2, 3]. Accordingly, the gaseous fuels generate almost exclusively thermal NO_x; with very little prompt NO_x and fuel NO_x, because gaseous fuel does not contain bound nitrogen, unlike liquid and solid fuels. The most important factors in thermal-NO_x emission can be expressed by the formula for NO formation [4]:

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

Where, K_1 and K_2 are constants, T is the temperature and t is the time. Because the flame temperature causes the separation of nitrogen in the air and combines it with oxygen and produces thermal NO which is highly temperature sensitive and in equation forms NO, as a function of temperature is involved in exponential and at temperatures above 1700 K every 40 degree increase in temperature will double production of NO [4]. If the technique can reduce the flame temperature it is effective to reduce NO_x formation [5, 6]. The indirect

effect of nitrogen oxide compounds can produce ozone and photochemical smog [7]. As a result, minimizing NO_x production Methods and knowledge in combustion is the most important thing. Because of the high activation energy of the thermal-NO Mechanism, due to the strong triple bond in the N₂-molecule, any scheme that suppresses peak temperatures will lower the NO output [7]. FGR is the most widespread technique in industry. Part of the flue gases is re-injected in the burner or mixed with the combustion air by means of a high-temperature fan and control system [4]. The major gases that cause back mixing fuel and air dilution and reduced concentrations of oxygen are species CO₂ and H₂O. The high heat capacity of these species increases thermal capacity of mixture, absorbing heat from the combustion and thus reduces the flame temperature and NO_x production [8, 9]. Although FGR method is a kind of dilution, but has economically advantages over dilution methods due to usage of gases of combustion. Also due to return of hot gases and the presence of active radicals in the combustion, thermal efficiency will be improved.

2. Experimental

In order to test the effect of flue gases recirculation on NO_x formation, a furnace has been designed and constructed. The main body of the furnace or combustion chamber is made of AISI316 steel in shape of hollow tube, 100 mm in diameter and 1000 mm in length formed that can tolerate high temperatures. Air needed for combustion at different equivalence ratios has been provided and the flow rate was controlled by an air valve. In order to assure good mixing after the initial mixing in the inlet pipe, the air and fuel enter venturi and then through a tube of 35 mm in diameter enters the combustion chamber. For more effectiveness of this methods reducing NO_x, Flue gases lost its heat in two stages; first in the main stacks and then in the suction line which a second heat exchanger was employed. The Schematic of the experimental set up has been illustrated in figure 1.

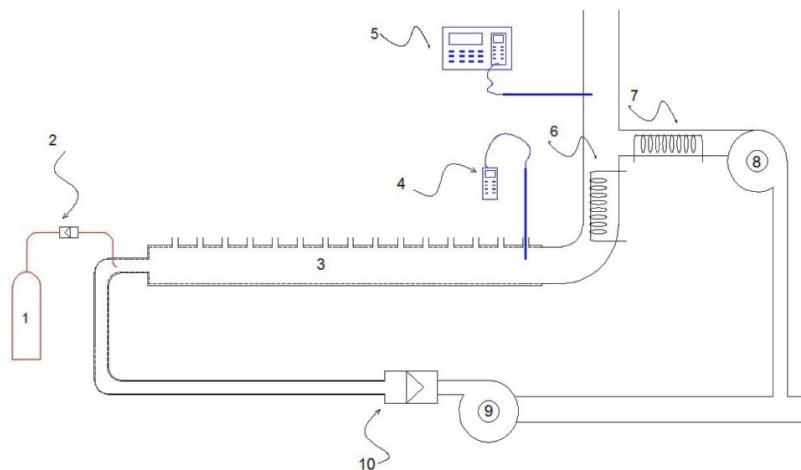


Figure 1: Experimental setup; 1, Propane; 2, Rota meter; 3, Test section; 4, Temperature sensor; 5, Gas analyser; 6, Heat exchanger; 7, Heat exchanger; 8, Suction fan; 9, Air fan; 10, Flow meter.

Suction of flue gases backs into the gas circuit by a valve on the fan, which is adjustable from 0 to 30 percent. Recycled gas enters combustion chamber with the inlet air after suction. Concentrations of species in flue gases are measured by the gas analysis system Testo 350 XL by putting sensors in the flue gases pipe. This device is capable of measuring the combustion efficiency. For measuring air flow, Lutron YK-2005AM device and for measuring flow rate of input fuel, SWPF-06A Rota meter is used. K-type sensor is also used for temperature measurements.

3. Numerical Simulations

In this study, Premix code of CHEMKIN II software package [10] is used. In simulations detailed mechanism of GRI-Mech3.0 [11] including 53 species and 325 chemical reactions has been used for the methane combustion. Profile mesh network scheme in simulation GRAD = 0.9 and CURV = 0.9 and the relative and absolute error criteria, respectively are RTOL = 1E-4 and ATOL = 1E-9. According to output of program the mode without recycle flue gas, mole fraction of the emissions is obtained according to the percentage of



various recycled gases that varies from 0 to 30 percent. This has been applied in the input code of Premix and its effect on the maximum flame temperature and NOx formation was evaluated. In the simulations the equivalence ratio varies from 0.7 to 1.3. Species available in the Table 1 have the most mole fraction in comparison with other species in the flue gases that is approximately 99.99% of the combustion products. As it can be seen from Table1 the mole fractions of H₂O, CO₂, N₂ gases have the largest percentage of emissions therefore in the second stage of simulations, the effect of these species only has been considered as the recycle gases. At this stage of the simulations, time saving has been accomplished, but its accuracy is less than simulations in first stage.

Table 1: Mole fraction of species contained in Flue gases.

ϕ	H ₂ O	O ₂	N ₂	CO ₂	CO	H
0.7	0.1108	0.05799	0.7432	0.08076	0.002475	0.000239
0.8	0.1281	0.03699	0.7381	0.09428	0.000702	0.000020
0.9	0.1413	0.01838	0.7296	0.1011	0.004639	0.000189
1	0.1526	0.004371	0.7201	0.1027	0.001320	0.000411
1.1	0.1496	0.004715	0.6965	0.08178	0.04177	0.004217
1.2	0.1467	0.003202	0.6781	0.07046	0.05991	0.008786
1.3	0.1528	0.000039	0.6656	0.06663	0.05668	0.001852

ϕ	O	OH	H ₂	NO
0.7	0.0007964	0.002942	0.0006874	0.00000834
0.8	0.0001282	0.001692	0.0001904	0.00003921
0.9	0.0003541	0.003161	0.001182	0.00005779
1	0.0002308	0.002852	0.003489	0.00009314
1.1	0.001080	0.005381	0.01440	0.00005370
1.2	0.001310	0.005379	0.02612	0.00004971
1.3	0.00001727	0.000616	0.05076	0.00003436

4. Results and Discussion

In Experimental and numerical study, equivalence ratio is varied from 0.7 to 1.3 and the FGR percent is variable from 0 to 30. To calculate the percentage of FGR the following relationship is used.

$$FGR \% = \frac{m_{recycle\ gas}}{m_{flue\ gas}} \quad (2)$$

Figs. 2, 3 show the simulations and experimental results of the effect of FGR on the flame temperature respectively for a range of equivalence ratio. The temperature measurements are taken at the distance of 600 mm from the beginning of the combustion chamber. It can be seen from figure 3 that with increasing FGR, the maximum flame temperature decreases. In the equivalence ratio 1, the temperature is higher than other equivalence ratios. Due to complete combustion of the Stoichiometry mode and the presence of oxygen is required for complete oxidation of fuel. In the presence of species of recycled gases, thermal mixing capacity increases. This reduces the heat caused by combustion and thus the flame and combustion chamber temperature will be reduced. With increasing FGR, oxygen mole fraction is reduced; leading to reduction in the heat release of combustion. Recycle gases with inlet air entering the combustion chamber. Flue gas recirculation in combustion chamber causes to pre-heat inlet air. Pre-heating the combustion air inlet is one of the methods of improving the quality of combustion and increase thermal efficiency. Generally, for pre-heating of inlet air heater is used in the inlet air path, but FGR technique has the advantage of using the flue heat for this purpose which will be saves energy consumption. Although pre-heating inlet air is one of the increasing the maximum flame and combustion chamber temperature factors, but due to the dilution effect of flue gases causes increase heat capacity and decrease combustion chamber temperature. Figure 4 and 5 shows the effect of FGR on the pre-heating inlet air and combustion efficiency. The temperature of recycle gas and air mixture in the inlet pipe is obtained by the thermometer.

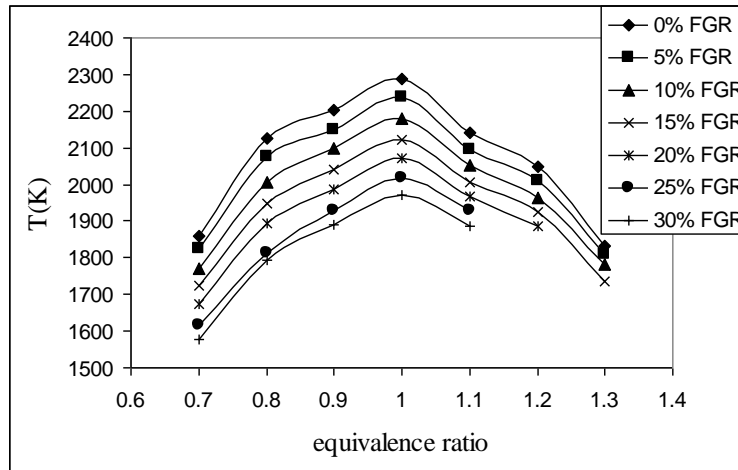


Figure 2: Effect of FGR on flame temperature (simulations).

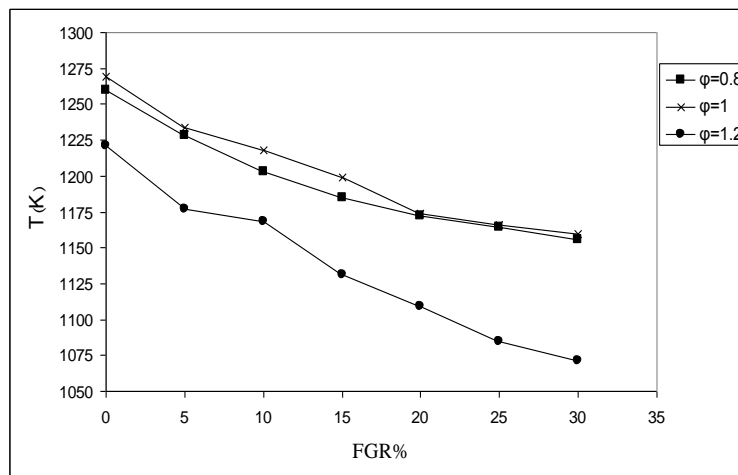


Figure 3: Effect of FGR on combustion temperature at a distance of 600 mm from the beginning of combustor (Experimental).

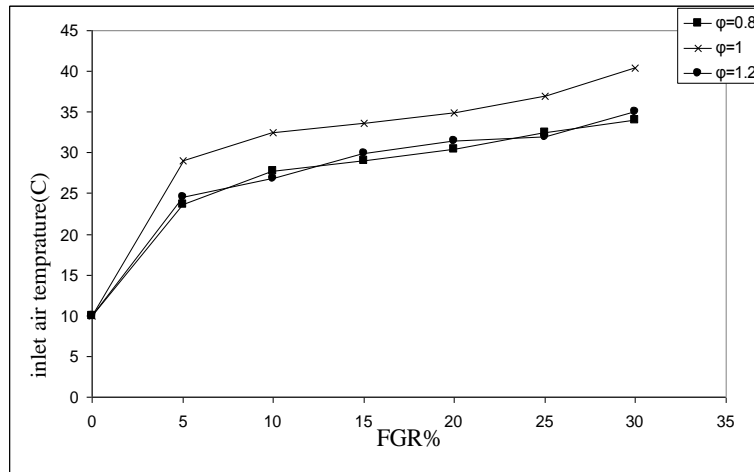


Figure 4: Effect of FGR on pre-heating of inlet air.

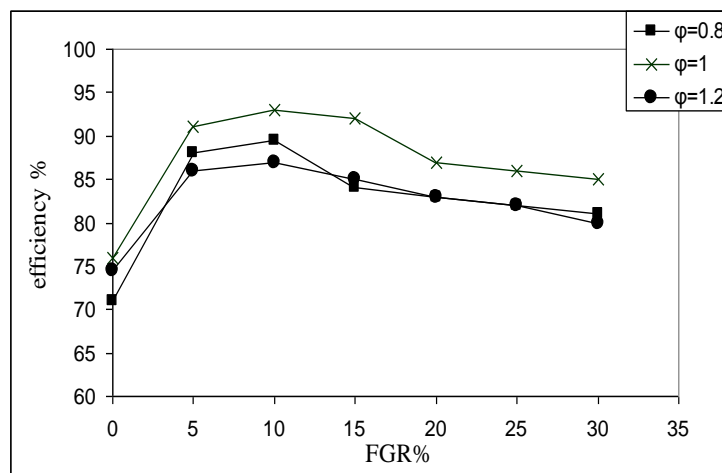


Figure 5: Effect of FGR on combustion efficiency.

It can be seen from figure 4, by increasing of recycled gases to the combustion chamber, inlet air temperature increases, which acts as pre-heating air. From figure 5 maximum efficiency is achieved with 10 percent recycled gas. Combustion efficiency depends on the



various parameters including equivalence ratio, combustion chamber temperature and pre-heating air. On the other hand with increasing FGR, the combustion temperature decreases, causing reduction of thermal efficiency. But inlet air pre-heating has higher effect than decrease of combustion chamber temperature. Figure 6 compares the simulations and experimental results of NO_x emissions for different equivalence ratios.

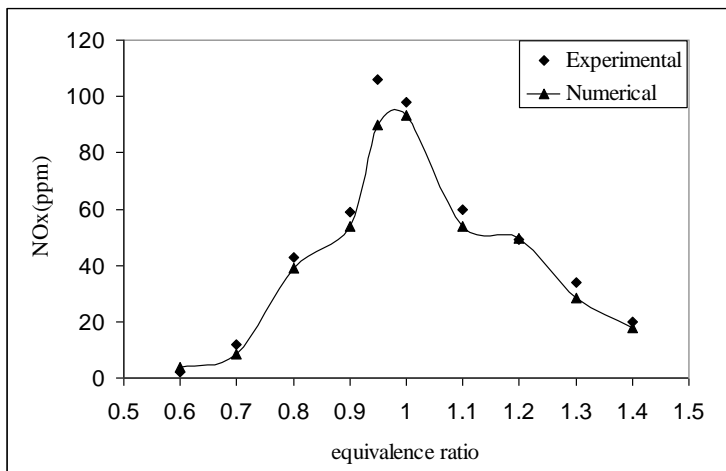


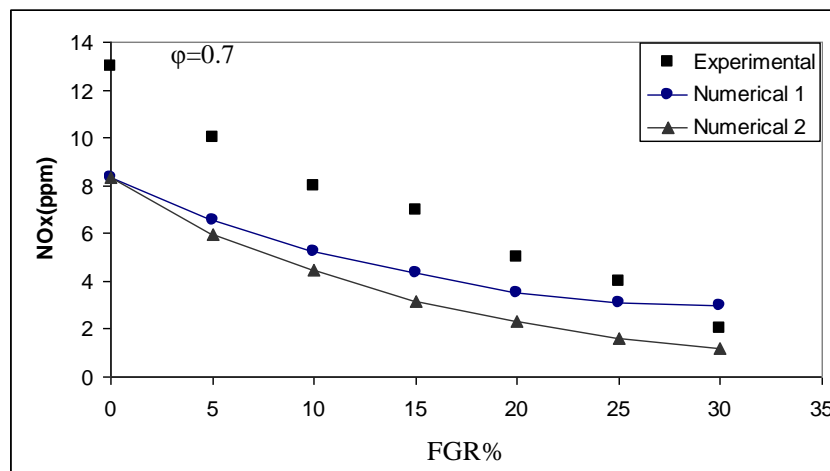
Figure 6: Effect of equivalence ratio on NO_x formation and comparing experimental and numerical results.

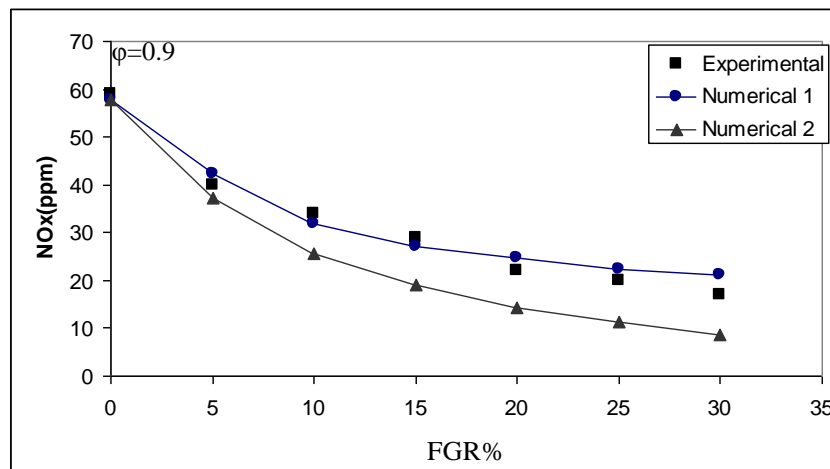
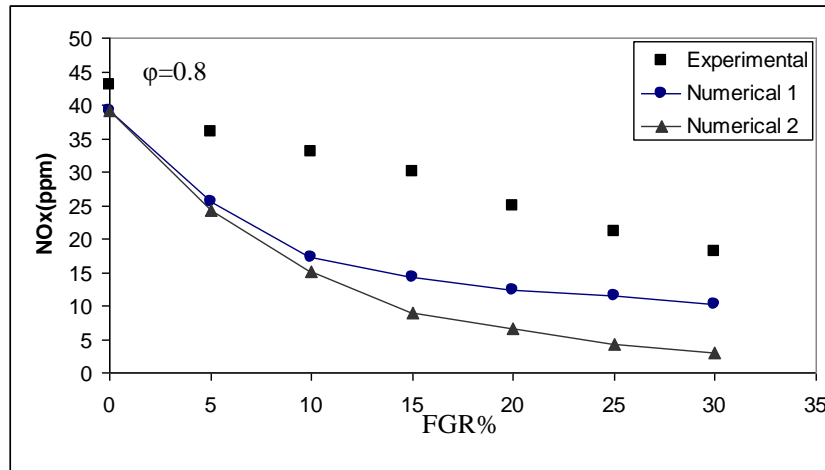
By increasing the equivalence ratio from 0.7 to near Stoichiometry mode, i.e. equivalence ratio of 0.95, NO_x emissions are increased. After this maximum value, by reaching fuel rich conditions and increasing equivalence ratio, NO_x is reduced. Cause of this change in NO_x emissions is the combustion temperature and oxygen and nitrogen concentrations in premixed mixture of input and also reduction of resident time. The Gas analyser has capability to measure NO and NO₂ and these are shown in the Table2. Since the NO₂ emissions are much smaller than NO emissions, therefore NO concentrations are considered as NO_x.

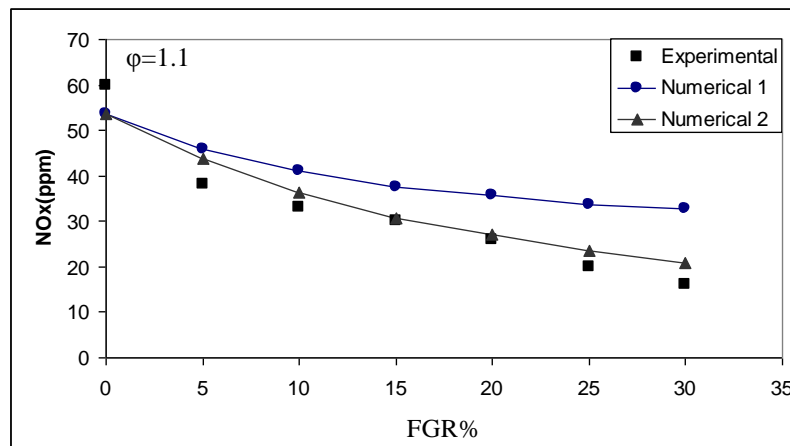
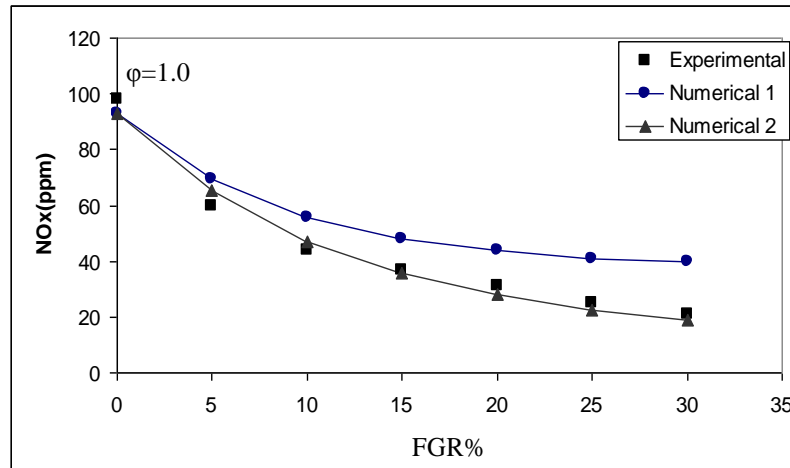
Table 2: Separation of NO_x emissions by NO and NO₂ in the premixed flame of methane – air (Experimental).

ϕ	NO (ppm)	NO ₂ (ppm)	NO _x (ppm)
0.7	13	0.3	13
0.8	42	0.5	43
0.9	58	0.6	59
1	96	2	98
1.1	59	0.8	60
1.2	49	0.2	49
1.3	34	0	34

Figure 7 shows the effect of flue gas recirculation on NO_x emission in range of equivalence ratios. The experimental results have been compared with both numerical results, considering all the species and only the important ones H₂O, CO₂, N₂ as a percent of FGR.







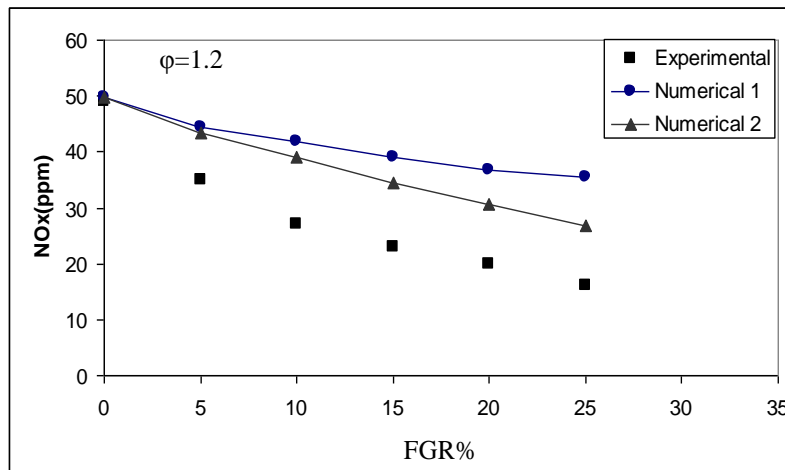


Figure 7: Effect of FGR on NOx emission and comparison of experimental and numerical results.

As it can be seen from figure 7, by increasing of FGR percent, NOx emissions are decreased. Since temperature is the main factor in NOx formation, all factors that reduce the maximum flame temperature and thus reduce the temperature of the combustion chamber are effective factors in NOx emissions reduction. Dilution effect of recycle gases, increases thermal capacity due to presence of mixed species with high thermal capacity, reduction of oxygen concentration in the inlet, increase of turbulence and decrease in resident time are the reasons of reducing the flame temperature and NOx emissions. Also by decreasing mole fraction of oxygen, the partial pressure is reduced which results decrease in NOx emission. Figure 8 shows the effect of recycled gases temperature on NOx emission. Recycled gases temperature is adjustable by using the heat exchangers in the main stack and the suction line.

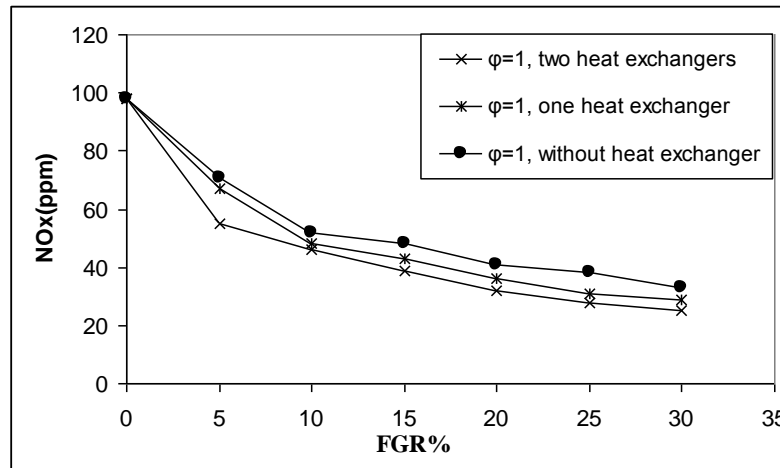


Figure 8: Effect of recycled gases temperature on NOx emission.

The level of NOx emission is higher in the state of using one heat exchanger, in the main stack than using both heat exchangers. This is because the inlet air gets cooler. Figure 9 shows the effect of equivalence ratio on NOx emission and comparison between present experimental and Cheng et al [12] experimental results. Figure 10 shows the effect of flue gas recirculation on NOx emission. The present numerical and experimental results are compared with Cho et al experimental data [7].

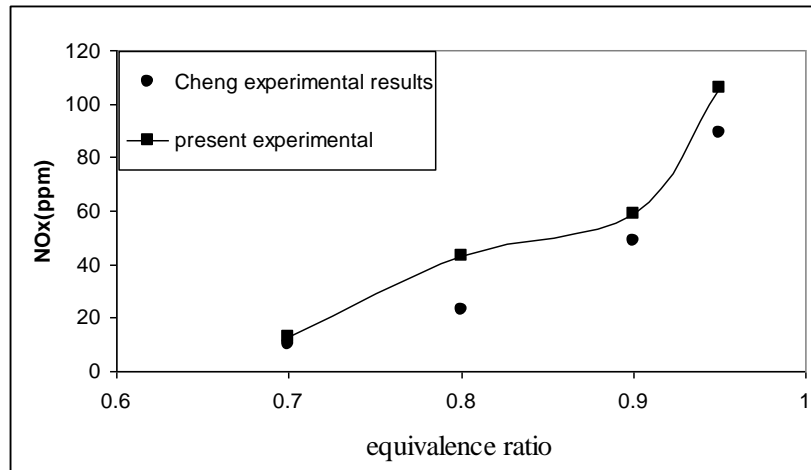


Figure 9: Comparison between present experimental and Cheng experimental results.

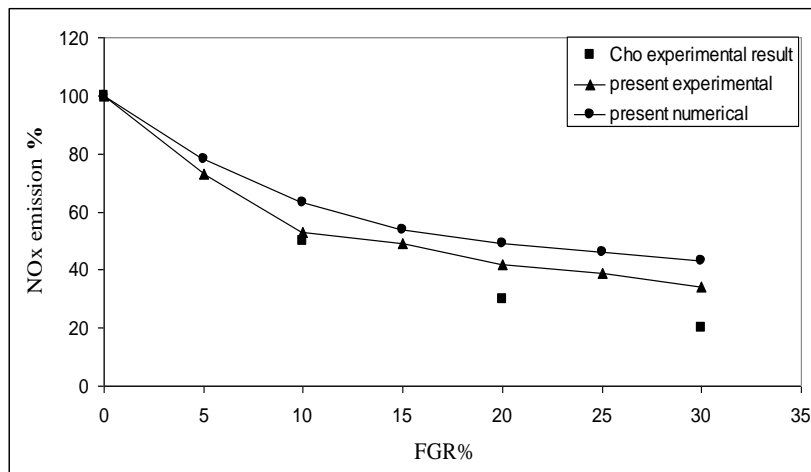


Figure 10: Comparison of present experimental and numerical results with Cho et al experimental data.

The present Simulation and experimental results show good agreement with Cheng [12] and Cho [7] experimental results.

Conclusion

- Flue gas recirculation into combustion chamber causes an increase in heat capacity and a decrease in oxygen concentration which reduces combustion temperature.
- Increasing the recycled gases to combustion chamber, the inlet air temperature increases and as the result the combustion efficiency also increases. Maximum efficiency in the combustion is achieved with 10 percent recycled gas.
- Maximum NO_x emissions are found to be at the equivalence ratio of 0.95.
- Experimental results at the equivalence ratio of 1.0 with 30% of FGR show 79 percent reduction in NO_x emission.

Increasing the recycled gas temperature, causes NO_x formation increases.

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