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NO_X FORMATION IN COMBUSTION OF NATURAL GASES USED IN TURKEY UNDER DIFFERENT CONDITIONS

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Abstract: The amount of NO_x produced during combustion is computationally investigated for methane, Russian-Shebelinka, Iran-South Pars, and Turkey-Tekirdağ natural gases in different operating conditions. The flames are laminar and stoichiometric premixed. The chemical kinetic mechanism used in the study consists of 59 reactions and 25 chemical species. A computer program (CREK) is used to compute mole and mass fractions of products. The highest and the lowest NO_x amounts are obtained during methane-air and Turkey-Tekirdağ natural gas-air combustions. Amounts of methane and nitrogen in the compositions of natural gases affect the amount of NO_x produced during combustion. The amount of NO_x increases with increasing reactant inlet temperature. Increased reactant total mass amount raises the amount of NO_x, but it causes mass fraction of NO_x to decrease in the products. Increasing humidity ratio of the burning air reduces the amount of NO_x.

Key words: Nitrogen oxide, Reaction mechanism, Natural gas combustion

TÜRKİYE'DE KULLANILAN DOĞAL GAZLARIN FARKLI KOŞULLARDA YANMASINDA NO_X OLUŞUMU

Özet: Yanma süresince üretilen NO_x miktarı, hesaplamalı olarak farklı işletme şartlarında metan, Rusya-Shebelinka, İran-Güney Pars, ve Türkiye-Tekirdağ doğal gazları için araştırılmıştır. Alevler laminar ve stokiometrik önkarışımlıdır. Çalışma içersinde kullanılan kimyasal kinetik mekanizma 59 reaksiyon ve 25 kimyasal bileşenden meydana gelmektedir. Ürünlerin mol ve kütle oranlarını hesaplamak için bir bilgisayar programı (CREK) kullanılmıştır. En yüksek ve en düşük NO_x miktarları metan-hava ve Türkiye-Tekirdağ doğal gazı-hava yanmalarında elde edilmiştir. Doğal gazların kompozisyonundaki metan ve azot miktarları yanma süresince oluşan NO_x'un miktarını etkilemektedir. NO_x miktarı artan ürün giriş sıcaklığı ile artmaktadır. Tepkimeye giren maddelerin toplam kütle miktarının artırılması, ürünler içindeki NO_x miktarını yükseltmekte fakat NO_x'un kütle oranının azalmasına neden olmaktadır. Yakma havasının artan nem oranı NO_x miktarını azaltmaktadır.

Anahtar kelimeler: Azot oksit, Reaksiyon mekanizması, Doğal gaz yanması

NOMENCLATURE

CREK	a computer program for calculation of combustion reaction equilibrium and kinetics in laminar or turbulent flow
EMV	total convective and diffusive reactant mass inflow rate to the control volume [$kg/m^3 \cdot s$]
ER	equivalance ratio
NO_x	nitrogen oxides
NO	nitrogen monoxide
NO_2	nitrogen dioxide
T_g	reactant inlet temperature [K]
k	rate coefficient [$kmol/m^3 \cdot s$]
w	humidity ratio [kg_{wv}/kg_{da}]

INTRODUCTION

Natural gas is used as fuel in power plants, vehicles and houses. Natural gas consists of methane (%85-95), ethane (%3-13), propane (%1-3), carbon dioxide (%0-4), nitrogen (%0.1-3), and other components (butane, pentane, hexane and water) (%1). Purified natural gas for the industrial aimed usage includes %99 methane. When hydro-carbon based fuels are especially compared according to their environment pollutants produced during their combustions, it is seen that natural gas is cleaner and increasingly preferred. The main pollutant for natural gas is NO_x. NO_x consists of NO (nitrogen monoxide) and NO_2 (nitrogen dioxide) and the amount of NO is higher.

The formation of NO_x during a combustion process occurs in three ways. These are thermal NO_x, fuel NO_x and prompt NO_x (EPA Technical Bulletin, 1999). Thermal NO_x is formed from the reaction of molecular nitrogen with oxygen atoms in the burning air at high temperatures over 1300 °C. It was defined by Zel'dovich as $N_2 + O \leftrightarrow NO + N$, $N + O_2 \leftrightarrow NO + O$, $N + OH \leftrightarrow NO + H$ reactions. Fuel NO_x results from oxidation of nitrogen's compounds and nitrogen in fuel. Prompt NO_x is formed from the reaction of carbon and hydrocarbon radicals with molecular nitrogen in the burning air in fuel-rich conditions.

Pollution produced during combustion and its amount depends on the type and the amount of fuel, properties of fuel and burning system, system operating procedures and meteorological conditions (Yolcu, 1997). The most important factors affecting the amount of NO_x are fuel composition, excess air, preheating temperature, and furnace temperature. Waibel reported that increasing preheating temperature causes NO_x emission more (Waibel, 1997). Each 3 °C increment in the temperature of the burning air in a premixed combustion system approximately raised 1 °C to the temperature of gas fuel mixed with the burning air (Alasfour, 1997). El- Sherif (El-Sherif, 1998) studied nitrogen oxides, flame structures, and burning velocities of different natural gases and reported that CARB standarded natural gas emitted NO_x lesser. Zevenhoven and Kilpinen (Zevenhoven and Kilpinen, 2004) concluded that increasing turbine inlet temperature increased NO_x emission. Turbiez et al. (Turbiez et al., 2003) experimentally studied stoichiometric premixed methane, methane-ethane, methane-ethane-propane, and natural gas flames and resulted that methane-ethane-propane and natural gas combustions exhibited similar characteristics by comparing mole fractions of O, H, OH, and HCO. Hırkalıoğlu (Hırkalıoğlu, 2005) reported that the NO_x emission distributions resembled to temperature distributions in chambers by using two different combustion chambers for combustions of methane, hydrogen and propane. NO_x causes acid rain, photo-chemical smog, and the depletion of the ozone (Mendioroz et al., 2006). It decreases the ability of blood to transport oxygen and minimizes the resistance against bacterial and viral infections in lung and makes negative effects on cells and the membrane of lung (Sağlık Vakfı, 2007). The present work investigates the amounts of NO_x produced during combustion processes of four different natural gases and how NO_x is varied in different operating conditions.

COMPUTATION

Combustion can theoretically be analyzed with the thermo-chemical equilibrium or the kinetic solution. In the equilibrium solution, it is assumed that there is infinite time for each fuel molecule to react with oxygen and reaction velocities are infinite. In the other words, combustion occurs in infinite small time interval. Besides, it can not be known that the system comes how fast to it's last situation because the reactivity of the system is not considered in the equilibrium case. It is not preferred to investigate combustion by the equilibrium solution because gases stand throughout a few seconds in the burning chamber during combustion process. Therefore, the reaction kinetic including a reaction mechanism and giving time-depended variations is important for the investigation of a real combustion process. In this study, it is used a computer program (CREK) to compute product moles and mass fractions in the kinetic solution. CREK (A Computer Program for Calculation of Combustion Reaction Equilibrium and Kinetics in Laminar or Turbulent Flow) presented by Pratt and Wormeck in 1976 calculate product concentrations by solving the equations of total mass and momentum conservation with respect to variables as velocity, pressure (Pratt and Wormeck, 1976). In the kinetic solution, algebraic equations are obtained by writing differential equations as finite-difference equations recognizing the problem for each node in grid after the solution area is divided into small areas in the shape of grid. Algebraic equations are used to calculate variables as temperature, pressure, concentration etc. through the solution area by utilizing the iterative or the matrix method for the beginning and limit conditions (Eyriboyun, 1997).

REACTION MECHANISM AND FUELS

It is not possible to know and observe reactions occurring in a real combustion process because the temperature of the combustion chamber is high, reactions are too fast and the zone of reaction is in a few millimeters. In theoretical studies, it is seen that reactions, and species, their numbers in reaction mechanisms used to analysis the combustion process for any fuel at certain conditions are different because combustion can not exactly solved. In the present study, the most appropriate reaction mechanism that fits to the limitations that CREK puts on the reaction types is selected from the literature. The chemical reaction mechanism and it's rate parameters are taken from El-Sherif's paper for natural gas-air combustions in 1998. The mechanism consists of 59 reactions and 25 chemical species (El-Sherif, 1998). Fuels used in this study are methane, Russia-Shebelinka (Dinca et al., 2006), Iran-South pars (Javanmardi et al., 2006) and Turkey-Tekirdağ (Arın and Akdemir, 2002) natural gases. The compositions of natural gases are given in Table 1. The reaction mechanism and its parameters are given in Table 2.

Table 1. Compositions of natural gases selected for the present study.

	Russia-Shebelinka	Iran-South Pars	Turkey-Tekirdağ	Methane
Methane (vol%)	96.2	97.4	92.6	100.0
Ethane (vol%)	1.2	0	0.5	0
Propane (vol%)	0.3	0	0.1	0
Carbon dioxide (vol%)	0.3	2.1	0.1	0
Nitrogen (vol%)	1.8	0	6.4	0
The remains (water, butane pentane, hexane etc. vol%)	0.2	0.5	0.3	0

Table 2. Parameters of rate coefficients and reactions in the kinetic mechanism of the combustion of natural gas-air. Rate coefficients are expressed as $k=AT^{B}\exp(-C/T)$ (El-Sherif, 1998).

No	Reactions	log *A*	*B*	*C*(K)
1	$OH + H_2 \leftrightarrow H_2O + H$	9.041	1.300	1825.0
2	$H + O_2 \leftrightarrow OH + O$	14.255	0.000	8450.0
3	$O + H_2 \leftrightarrow OH + H$	5.255	2.500	3300.0
4	$H + O_2 + M \leftrightarrow HO_2 + M$	20.574	-1.72	0.0
5	$H + HO_2 \leftrightarrow OH + OH$	14.342	0.000	710.0
6	$H + HO_2 \leftrightarrow O + H_2O$	12.698	0.000	710.0
7	$H + HO_2 \leftrightarrow H_2 + O_2$	13.334	0.000	280.0
8	$OH + HO_2 \leftrightarrow H_2O + O_2$	13.255	0.000	0.0
9	$O + HO_2 \leftrightarrow OH + O_2$	13.301	0.000	0.0
10	$H + H + M \leftrightarrow H_2 + M$	18.000	-1.00	0.0
11	$H + OH + M \leftrightarrow H_2O + M$	22.204	-2.00	0.0
12	$H + O + M \leftrightarrow OH + M$	16.792	-0.60	0.0
13	$OH + OH \leftrightarrow O + H_2O$	12.760	0.000	390.0
14	$OH + CO \leftrightarrow CO_2 + H$	7.176	1.300	-385.0
15	$O + CO + M \leftrightarrow CO_2 + M$	15.732	0.000	2300.0
16	$H + CO + M \leftrightarrow CHO + M$	14.698	0.000	755.0
17	$CHO + O_2 \leftrightarrow HO_2 + CO$	13.518	-0.40	0.0
18	$CHO + H \leftrightarrow H_2 + CO$	14.079	0.000	0.0
19	$CHO + OH \leftrightarrow CO + H_2O$	14.000	0.000	0.0
20	$CHO + O \leftrightarrow OH + CO$	13.477	0.000	0.0
21	$CH_2O + H \leftrightarrow CHO + H_2$	4.000	3.000	700.0
22	$CH_2O + OH \leftrightarrow CHO + H_2O$	13.477	0.000	600.0
23	$CH_2O + O \leftrightarrow CHO + OH$	13.255	0.000	1540.0
24	$CH_4 + H \leftrightarrow CH_3 + H_2$	4.342	3.000	4400.0
25	$CH_4 + OH \leftrightarrow CH_3 + H_2O$	7.204	1.830	1400.0
26	$CH_4 + O \leftrightarrow CH_3 + OH$	7.079	2.100	3840.0
27	$CH_3 + O \leftrightarrow CH_3O + H$	13.845	0.000	0.0
28	$CH_3 + O \leftrightarrow CHO + H_2$	14.146	0.000	860.0
29	$CH_3 + CHO \leftrightarrow CH_4 + CO$	12.000	0.000	0.0
30	$CH_3 + HO_2 \leftrightarrow CH_4 + O_2$	12.301	0.000	0.0
31	$CH_3 + HO_2 \leftrightarrow CH_3O + OH$	12.301	0.000	0.0
32	$CH_3O + H \leftrightarrow CH_2O + H_2$	13.698	0.000	0.0
33	$CH_3O + OH \leftrightarrow CH_2O + H_2O$	13.397	0.000	0.0
34	$CH_3O + O \leftrightarrow CH_2O + OH$	13.397	0.000	0.0
35	$CH_3 + CH_3 + M \leftrightarrow C_2H_6 + M$	14.447	-0.40	0.0
36	$C_2H_6 + H \leftrightarrow C_2H_5 + H_2$	2.740	3.500	2620.0
37	$C_2H_6 + OH \leftrightarrow C_2H_5 + H_2O$	6.549	2.120	439.3
38	$C_2H_6 + O \leftrightarrow C_2H_5 + OH$	7.953	1.920	2864.0
39	$C_2H_5 + O_2 \leftrightarrow C_2H_4 + HO_2$	12.301	0.000	2510.0
40	$C_2H_5 + H \leftrightarrow CH_3 + CH_3$	13.681	0.000	0.0

41	$C_2H_5 + O \leftrightarrow C H_3 + C H_2O$	13.698	0.000	0.0
42	$C_2H_5 + C_2H_5 \leftrightarrow C_2H_4 + C_2H_6$	14.146	0.000	0.0
43	$C_2H_4 + O \leftrightarrow CH_3 + CHO$	13.301	0.000	1300.0
44	$C_2H_4 + OH \leftrightarrow CH_3 + CH_2O$	13.602	0.000	760.0
45	$C_2H_4 + OH \leftrightarrow C_2H_3 + H_2O$	13.845	0.000	1516.0
46	$C_2H_4 + H \leftrightarrow C_2H_3 + H_2$	14.176	0.000	513.0
47	$C_2H_3 + H \leftrightarrow C_2H_2 + H_2$	13.301	0.000	0.0
48	$C_2H_3 + O_2 \leftrightarrow C_2H_2 + HO_2$	12.000	0.000	0.0
49	$C_2H_2 + O \leftrightarrow C H_2 + CO$	8.612	1.500	854.0
50	$C H_2 + O_2 \leftrightarrow C O_2 + H_2$	14.000	0.000	1864.0
51	$N_2 + O + M \leftrightarrow N_2O + M$	13.740	0.000	10770.0
52	$N_2O + O \leftrightarrow NO + NO$	14.851	0.000	14100.0
53	$N + O_2 \leftrightarrow NO + O$	9.806	1.000	3162.0
54	$N_2O + H \leftrightarrow N_2 + OH$	13.880	0.000	7654.0
55	$N_2 + O \leftrightarrow N + NO$	14.414	0.000	38390.0
56	$N + OH \leftrightarrow H + NO$	11.799	0.500	0.0
57	$NO + O + M \leftrightarrow NO_2 + M$	17.000	0.000	0.0
58	$NO + HO_2 \leftrightarrow NO_2 + OH$	15.146	0.000	705.0
59	$NO_2 + O \leftrightarrow NO + O_2$	10.763	0.500	0.0

RESULTS AND DISCUSSION

Mass fractions of NO_x produced during the combustions of fuel-air mixtures are obtained by changing equivalence ratio (ER) from 0.6 to 1.4 for a constant reactant inlet temperature (T_g=298 K) and a constant reactant total mass amount (EMV = 0.01 kg/m^3·s) entering to the control volume via the convection and the diffusion at 1 atm and given in Fig 1. EMV also sets the time of reaction. For a certain reaction chamber, larger EMV value means short reaction time and vice versa. Reactant total mass equals to the total of masses of fuel and air that is necessary to burn fuel. NO_x reach to the maximum at ER=0.88 for all the fuels. NO_x must be maximum at ER=1 (El-Sherif, 1998). The results are in general agreement with the literature within a discrepancy of 12%. This discrepancy is due to CREK's calculation sensitivity and its the limitations that CREK puts on the reaction types entered in the data file.

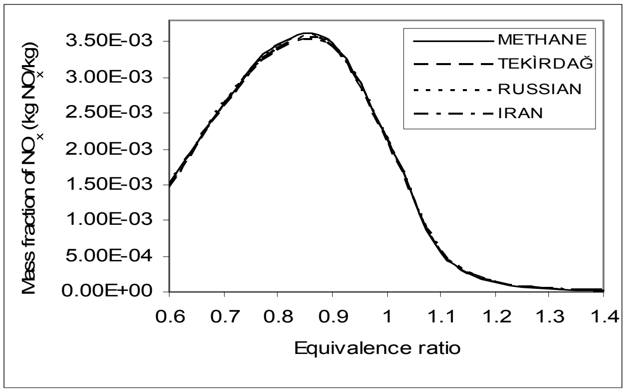

Figure 1. Computed mass fraction of NO_x with equivalence ratio at T_g=298 K, EMV=0.1 kg/(m^3·s).

There is no considerable difference among NO_x mass fractions. There is NO_x difference of 2% between Methane and Turkey-Tekirdağ natural gas-air combustions which have the highest and lowest NO_x emissions. Amounts of nitrogen and methane in natural gas have an important effect over the level of NO_x emission. Especially, high methane composition of fuels increases the reaction temperature and increasing temperature causes to thermal NO_x to form more. Although methane composition of Iran gas is higher than that of Russia gas, NO_x emitted during the combustion of Russian gas is higher because of Russian gas's nitrogen composition. The lowest NO_x emission is obtained during the combustion of Tekirdağ`s gas. Even though Tekirdağ`s natural gas has the highest nitrogen composition, its low methane composition causes the reaction temperature to decrease and decreasing reaction temperature lowers thermal NO_x amount to form.

Reactions of prompt NO_x formation are neglected to simplify the reaction mechanism by El-Sherif. It does not make a considerable effect on total NO_x formation because prompt NO_x formation occurs in the rich mixture zones and does not change with temperature. In this study, it is supposed that NO_x forming during combustion is total of NO_x forming via thermal and fuel NO_x formation. Thermal NO_x is dominant in total NO_x. The gas compositions that have volumetric percentages under 1 are neglected in computations. Besides, El-Sherif assumed the hydrocarbon compositions higher than C_2H_6 to be C_2H_6 for simplifying the complexity of the kinetics.

For ER=1 and T_g=298 K, when the reactant total mass amount (EMV) is increased, the amount of NO_x increases for the combustions of all the fuels as shown in Fig. 2. Fig. 3 shows that NO_x mass fraction does not show any variation with low EMV values (<0.1 kg/m^3·s) but mass fractions of NO_x decreases for EMV values higher than 0.1 kg/m^3·s. This decrease arises from high reactant mass amount entering to the burning chamber and some of hydrocarbons leaving the chamber without combusting completely because high reactant mass flow means short reaction time. Mass fractions of combustion products of Russian`s gas for EMV values of 0.1, 10 and 100 kg/m^3·s are given as a sample in Table 3. When mass fractions are compared, it is seen from their increasing mass fractions versus increasing EMVs that some amount of hydrocarbons as CH_4, C_2H_6 etc. do not burn completely.

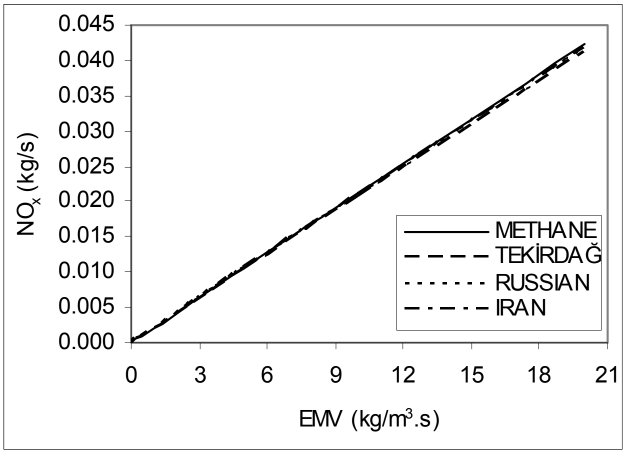

Figure 2. NO_x amount versus reactant total mass amount at ER=1, T_g=298 K.

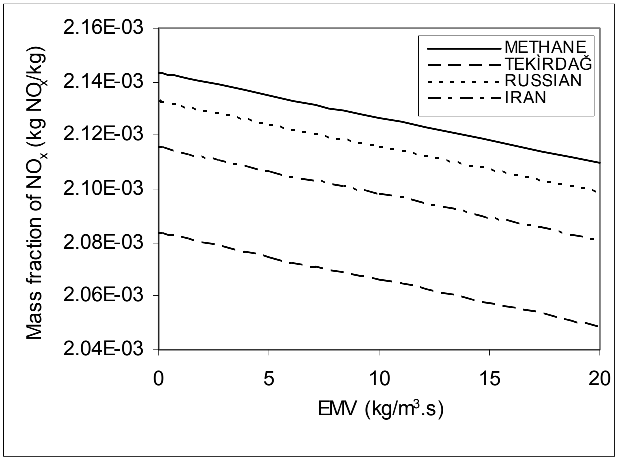

Figure 3. Mass fraction of NO_x as a function of EMV at ER=1, T_g=298 K.

NO_x emissions increases when the reactant inlet temperature is increased at EMV=0.01 kg/m^3·s and ER=1. The emissions are shown in Fig. 4 and Fig. 5. There is a 25%-increment between mass fractions of NO_x produced during the combustions for 223 K and 323 K inlet NO_x emissions increases when the reactant inlet temperature is increased at EMV=0.01 kg/m^3·s and ER=1. The emissions are shown in Fig. 4 and Fig. 5. There is a 25%-increment between mass fractions of NO_x produced during the combustions for 223 K and 323 K inlet temperatures. Altought high inlet temperature is desired to raise the productivity of the system, increased inlet temperature causes the temperature of the reaction to raise. Increasing reaction temperature increases the amount of NO_x forming via thermal NO_x formation. The effect on NO_x formation of increasing reactant inlet temperature for different reactant inlet masses is given in Fig. 6 for Russian-Shebelinka natural gas-air combustion. Mass fraction of NO_x decreases with increasing reactant inlet mass at raising reactant inlet temperature. The decrease is 1.1% between 0.01 kg/m^3·s and 10 kg/m^3·s. The same effect is seen for the other gas combustions as well. Fig. 7 shows that increasing reactant inlet temperature raises product outlet temperature up for EMV=0.01 kg/m^3·s, ve ER=1.

Table 3. Mass fractions of combustion products of Russian`s gas for different EMV values.

Products	**EMV=0.1 kg/m^3·s**	**EMV=10 kg/m^3·s**	**EMV=100 kg/m^3·s**
N2	.72376E+00	.72377E+00	.72376E+00
CH2	.26685E-14	.26621E-12	.26664E-11
CH2O	.76164E-10	.54668E-08	.54319E-07
C2H3	.83062E-13	.83144E-11	.83840E-10
C2H5	.31241E-11	.31231E-09	.31146E-08
C2H6	.86893E-12	.86855E-10	.86539E-09
O	.12135E-03	.12145E-03	.12227E-03
OH	.17797E-02	.17805E-02	.17871E-02
HO2	.66444E-06	.66524E-06	.67206E-06
H	.13900E-04	.13902E-04	.13916E-04
H2	.26203E-03	.26196E-03	.26141E-03
C2H4	.92799E-13	.92858E-11	.93372E-10
CH3	.18209E-09	.18194E-07	.18076E-06
CH4	.20672E-09	.20657E-07	.20537E-06
CO	.90745E-02	.90728E-02	.90584E-02
CO2	.13771E+00	.13771E+00	.13773E+00
C2H2	.15893E-12	.15905E-10	.16003E-09
O2	.52775E-02	.52847E-02	.53459E-02
H2O	.11987E+00	.11987E+00	.11987E+00
CH3O	.43362E-14	.43355E-12	.43369E-11
N2O	.14914E-06	.14924E-06	.15004E-06
CHO	.79196E-09	.79198E-09	.79223E-09
N	.70405E-08	.70400E-08	.70345E-08
NO	.21318E-02	.21149E-02	.19721E-02
NO2	.54472E-06	.54065E-06	.50620E-06

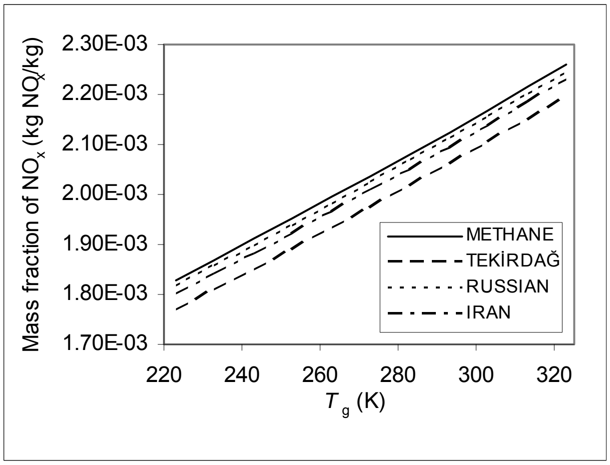

Figure 4. Mass fraction of NO_x with reactant inlet temperature at ER=1, EMV=0.01 kg/m^3·s.

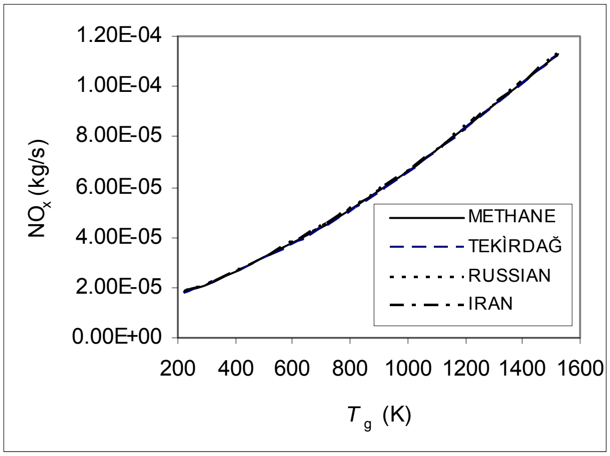

Figure 5. Amount of NO_x with reactant inlet temperature at ER=1, EMV = 0.01 kg/m^3·s.

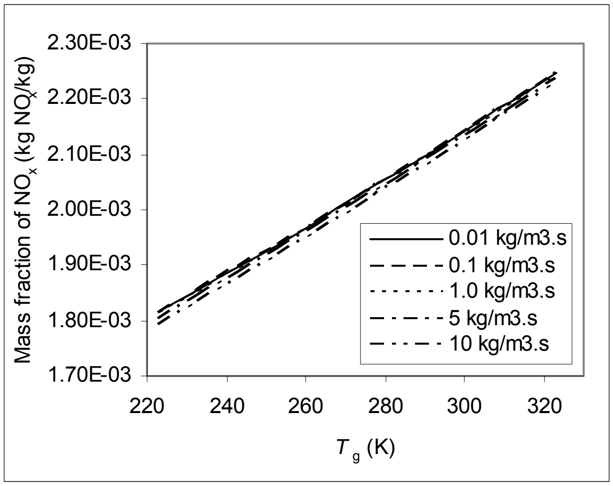

Figure 6. Mass fraction of NO_x with reactant inlet temperature at different EMV values and ER=1 for Russian natural gas-air combustion.

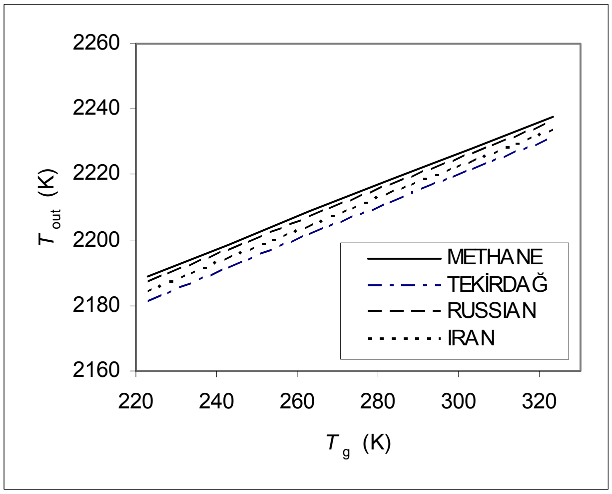

Figure 7. Product outlet temperature versus reactant inlet temperature for EMV=0.01 kg/m^3·s, ER=1.

NO_x emissions are examined by changing the humidity ratio from 0.006 to 0.022 kg_{wv}/kg_{da} at T_g=303 K, EMV = 0.01 kg/m^3·s and ER=1. The dry and wet thermometer temperatures and humidity ratios (w) of Zonguldak are taken into consideration for all the computations. When the humidity ratio of the burning air is increased, NO_x decreases for all the fuels. Variation in the amount of NO_x for the lowest and the highest humidity ratios (0.006 and 0.022 kg_{wv}/kg_{da}) is 13%. Increment of humidity in the burning air reduces the temperature of the reaction because both the humidity has highly the ability of holding the heat and it prevents the flame to feed by oxygen by wrapping as a cover over the flame. Thus, decreasing temperature of the reaction causes NO_x forming via thermal NO_x formation to decrease. NO_x emitted with respect to the humidity in the burning air is shown in Fig. 8.

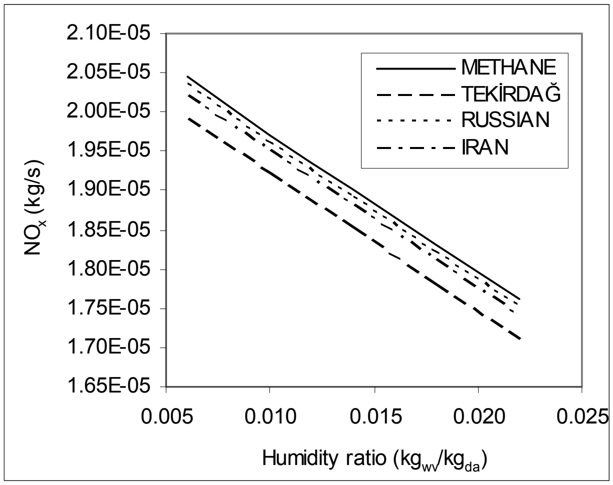

Figure 8. NO_x as a function of humidity ratio at ER=1, T_g=303 K and EMV = 0.01 kg/m^3·s.

CONCLUSION

The concentration of NO_x is computed for laminar premixed flames of methane, Iran-South Pars, Russian-Shebelinka and Turkey-Tekirdağ natural gases at 1 atm, 298 K and different ER from 0.6 to 1.4. The maximum mass fraction of NO_x for all the gases is at ER=0.88. The computed amount of NO_x is in a good agreement with the literature and discrepancy is 12%. There is variation of 2% between the lowest and highest mass fractions. The compositions of methane and nitrogen of natural gases affect the amount of NO_x. When EMV is increased, the amount of NO_x also increases for all the fuels but mass fractions of NO_x begin to decrease after 0.1 $kg/m^3 \cdot s$ because some of hydrocarbons in fuels do not burn. When the reactant inlet temperature is increased, the mass fractions of NO_x increases because increasing reaction temperature increases the formation of thermal NO_x. When the humidity ratio of the burning air is increased, the mass fractions of NO_x decreases because decreasing reaction temperature decreases the formation of thermal NO_x. The variation between mass fractions of NO_x forming during combustion for the lowest and highest humidity ratios is 13%. NO_x amount forming via thermal NO_x formation composed 85-90% of total NO_x amount produced during gas-air combustion. The discrepancies are caused by CREK's calculation sensitivity and reaction type limitations.

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