EXPERIMENTAL AND NUMERICAL SIMULATION ON NO_x EMISSION IN LIQUID FUEL SPRAY FLAMES

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Abstract
In this paper production and emission of *NOx* pollutant in a cylindrical furnace liquid fuel, for various angles and patterns of fuel spray was studied through the experimental and numerical method. Pollutants measurement has been done for fuel spray angles 45°, 60° and 80°, spray pattern of hollow and solid cone. Numerical simulation of two phase flow and combustion modelling for pollutants formation are done with Fluent6.32 software. Comparison of numerical and experimental results shows very good agreement. The results show that by increasing in spray angle NO_x emission increases. When the spray angle increases, the diameter of exhausted fuel particles from nozzle declines, which as a result the contact between fuel and air rises and also the air-fuel mixing increases. Therefore with increasing in the mentioned spray angle, a more perfect combustion happens and the maximum flame temperature increases. Also, the concentration of NO_X which is affected by maximum temperature increased.

Introduction
Combustion processes, usually lead to production of pollutants to the environment. In optimization of these processes not only the combustion efficiency and reduction fuel consumption, but also the environment and reducing certain pollutants should be considered. For this purpose, extensive researches have been done to save energy and achieve high efficiency and control and reduce pollution caused by combustion. A significant proportion of major air pollutants are produced from the combustion of liquid fuel. Liquid fuel is widely used in the boilers, industrial furnaces and gas turbine combustion chambers.

Sulfur oxide, carbon monoxide, soot and nitrogen oxides are most important contaminants which spread in the liquid fuel flames. Produced pollution by nitrogen oxides has remained harmful effects on human health and the environment and also plays an important role in the formation of acid rain, chemical smog and ozone layer hole. Laboratory measurements play an important role in engineering researches and because of the complex combustion process, numerical techniques and mathematical modeling of combustion process still is not fully reliable and laboratory results are needed. In the other hand because of special experimental condition and sensitive measurement devices, the laboratory results should be verified.

Liquid fuels are categorized to light and heavy fuels depending on the amount of carbon. Liquid fuel burners are usually non-premixed and the fuel and oxidizer are mixed in the combustion chamber. Gas fuels are widely categorized as premixed or non-premixed. In premixed burner smaller and stronger flame is produced, in comparison with the non-premixed type. Flame adiabatic temperature is higher and radiation heat transfer and also the rate of the formation of nitrogen oxides are increased. But in non-premixed type, flame is taller, the temperature at the center of flame and NO_x production are reduced [1]. Although the chemical processes of the both premixed and non-premixed combustion are the same but

additional physical processes regarding to the non-premixed combustion (evaporation and mixing) are needed. For example, overall mixing can be stoichiometric, but there are some areas in combustion chamber with large or low amount of fuel. This aspect of the non-premixed combustion can cause to more complex formation of pollutants [2].

Liquid fuel combustion is controlled by the known parameters such as atomization of the jet liquid fuel, droplets fuel spray evaporating, mixing of fuel and oxidizer. Speed, angle and pattern of fuel spray can be controlled by the burner nozzle. Very extensive researches have been done for the effect of the nozzle design, working conditions and fuel properties on the combustion characteristics and on spraying the hydrocarbon fuels [3-5]. Most conducted research has focused on the nozzles used in the combustion gas turbines and boilers. In gas turbine combustion, getting the high thermal efficiency and very small emissions of pollutants are very important. By increasing the turbine inlet temperatures it can reach to higher efficiency and combustion chamber design should be improved for lower combustion pollutants. In liquid fuel combustion in the gas turbine, there is the problem of the formation of soot and NO_x emissions and physical phenomena such as fuel atomization or evaporation, droplets dispersion, structure of spray and mixing with air, have widely effected on combustion.

In the past, research was often performed based on techniques and methods, to improve combustion efficiency and quality of atomization. Improvement of the burner efficiency requires the study of spray nozzle characteristics. Recently, most research is focused to review and to study spray characteristics such as length of segregation, spray angle and droplet size and distribution in the burner nozzle [6-7].

The purpose of this research is to study the production and emission of air pollutants, which produced from liquid fuel burner and to consider fuel spray angle effects on the emissions. In this study, two different methods as laboratory measurement and numerical simulations are used. To calculate the amount of NO_x pollutant, two mechanisms as thermal *NO* and fuel *NO* are employed and the obtained results are compared in two methods.

Case study and geometry of liquid fuel furnace

Laboratory furnace which is used in this study, includes a horizontal cylindrical with 1800 *mm* length and ratio of *L/D=5.625*. Inner surface of furnace is completely smooth to prevent resistance of hot gases movement. Furnace body is made from *AISI316* steel to resist high temperatures. Around the furnace is covered by high temperature thermal insulation with the proper thickness so that the heat transfer from the furnace body can be almost considered zero. For measuring the temperature and combustion gas concentration in different parts of the furnace including the body hull and the chimney, some places are embedded with the cap to get sample when it is necessary. Schematic experimental system is shown in Figure 1.

Three commercial nozzle spray angles of 45, 60 and 80 degrees with two spray patterns as hollow cone spray and solid cone spray are selected for testing. Furnace burner with maximum power of 240 *W* is pressure type and fuel inlet pressure to the nozzle equals to 1.5 *MPa*. Fuel and air flow rate entering to the combustion chamber are adjustable by the fuel pump and air throttle valve. Mixing fuel and air is provided with intake air swirl. Fuel is injected into the furnace through the nozzle. Burner characteristic are presented in Table 1. Tests are done for the diesel fuel with input temperature 40^oC and viscosity is 1.5-5.1 centistokes. Other fuel profile is given in Table 2. Laboratory furnace is designed in such a way that the fuel spray nozzle can be easily replaced. Since the surface roughness of

components inside nozzle is effective to atomize the fuel particles, those nozzles with available calibration and verification evidence for their performance are used.

All measurements are performed after the furnace temperature reaches steady state because the temperature changes can cause to alter the fuel concentration and viscosity and these factors play an important role on size of fuel droplets. During the tests the inlet temperatures of air and fuel are under control and are kept constant. A sampling device has been located within 160 cm from the furnace vent in order to analyze exhaust gas in the chimney. Flow of produced combustion gases such as CO_2, CO, NO_x, and combustion efficiency are measured every 5 minutes by using a gas analyzer system (Testo*350XL*).

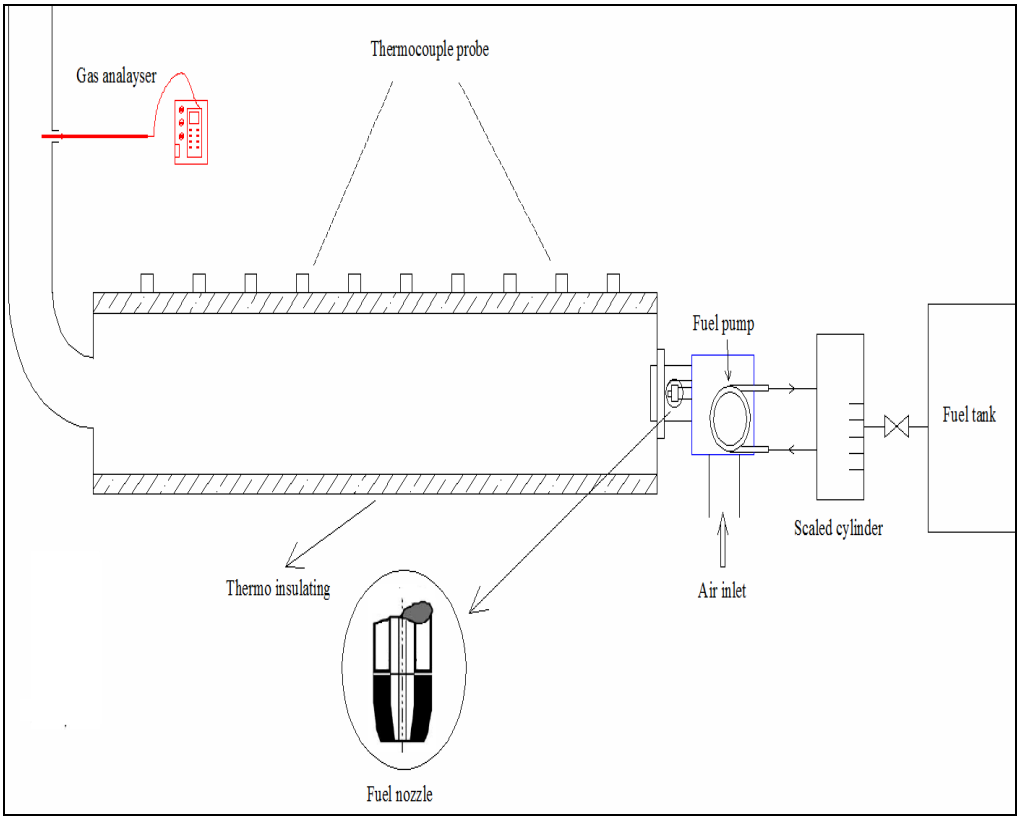


Figure 1. Schematic of experimental furnace for liquid fuel.

Table 1. Features of burner.

Power source	AC 220V / 50 – 60 Hz
Motor	240 W
Oil pump type	Gear pump
Pump pressure	1.5 Mpa
Nozzle fuel rate	0.85 – 1.25 gal/hr
Spray angle	45, 60, 80 degree

Table 2. Properties of gasoline at a temperature of 295 K.

Density	830 kg/m^3
C_p	2005 J/Kg*K
Vaporization Temperature	373 K
Boiling Point	462 K
N_2 Percentage	0.09 %

Governing equations

Starting point for computing the flows without combustion is to solve Navier – Stocks equations. For the flows including with the heat transfer, the energy equation is added. Completing of these conservative equations is equation of state and fluid properties characteristics. Combustion flows are included of the release of thermal energy because of conversion of chemical species. In this kind of combustion flows, the combustion process modeling and adding to the Navier–Stocks equations are needed.

Two-phase flow Modeling

In many practical combustion processes, fuel is solid or liquid and during the combustion, firstly the fuel is converted to the gas phase and then is burned by oxidation gas. Liquid fuel combustion is usually done by injecting liquid fuel into the gas phase combustion environment. Turbulence within the liquid flow is created inside the injector. This turbulent fluid flow gets out of the nozzle in the form of complex mix of strings and then crushes into small drops and appears as dense clouds of droplets that launches through the gas into the flame zone. Heat transfer to drop, increases the vapor pressure and therefore the fuel evaporates into the gas, so burning of gas phase is began. Non-premixed flame surrounds the group drops or droplets and eventually fuel vapor burns. This set of processes is called combustion spray.

By assuming that spherical particles of liquid fuel is dispersed in the gas phase and their hits due to their quick evaporation is negligible, in the Lagrangian System, continuity and energy conservation equations governing to the particles, are written as follows:

$$\frac{dd_p}{dt} = -\frac{C_b}{2d_p}\left(1 + 0.23\,\mathrm{Re}^{1/2}\right) \tag{1}$$

$$\frac{dT_p}{dt} = \frac{6k\left(2 + 0.6\,\mathrm{Re}^{1/2}\,\mathrm{Pr}^{1/3}\right)\left(T_\infty - T_P\right)}{\left(\rho_P d_p^2 C_{P,P}\right)} \tag{2}$$

In equation (1), d_p is drop diameter C_b is evaporation constant which is function of physical properties of the environment and fuel that is determined as follows:

$$C_b = \frac{8k}{\rho_l C_p}\ln\left[1 + C_p / L\left(T_\infty - T_P\right)\right] \tag{3}$$

In this equation k and C_p are thermal conductivity coefficients, and the specific heat in constant pressure for gas mixtures, respectively. T_∞ , T_p and L are gas temperature, the drop temperature and the latent heat of vaporization of fuel respectively. By using equation (1), the particle diameter change rate, after reaching to the boiling temperature, and from equation (2) the temperature change rate of the fuel particles can be calculated [8].

Combustion modeling

In this study chemical formula for gasoline is assumed to be $C_{16}H_{29}$ and one step Magnussen-Hiertager model with eddy dissipation is used for combustion modeling [9]. In this simulation firstly, the liquid fuel converts into a gas type and then combustion is performed. Evaporation of fuel particles is started when the particles temperature reaches to the boiling temperature, and will be continued until all their mass becomes finish. Gas fuel is done in one step and according to the selected Magnussen-Hiertager combustion model; the effective factor to the reaction rate is the flow dynamics. Chemical reaction between oxygen and gasoline is written as follows:

$$C_{16}H_{29} + 23.25O_2 \longrightarrow 16CO_2 + 14.5H_2O\,. \tag{4}$$

Nitrogen oxide modeling

To estimate *NO*, the solution of the mass transport equation (Y_{NO}) is required. This equation is solved after determining the main flow field and their main species. Mass transport equation for *NO* is written as follows, which includes influence convection term, production and consumption of *NO*.

$$\rho\frac{\partial Y_{NO}}{\partial t} + \rho u_i\frac{\partial Y_{NO}}{\partial x_i} = \frac{\partial}{\partial x_i}\left(\rho D\frac{\partial Y_{NO}}{\partial x_i}\right) + S_{NO} \tag{5}$$

Where source term (S) in this equation is calculated according to three forms mechanism *NO*: thermal NO, prompt *NO* and fuel *NO*. Since in liquid fuels combustion the amount of prompt *NO* compared with the other two types are almost neglected, in this study only thermal *NO* and fuel *NO* are calculated. Thermal *NO*, which is formed at high temperatures by nitrogen oxidation contained in combustion air, is expressed using developed Zeldovich mechanisms [10].

$$O + N_2 \xleftrightarrow{k_{\pm 1}} NO + N \tag{6}$$

$$N + O_2 \xleftrightarrow{k_{\pm 2}} NO + O \tag{7}$$

$$N + OH \xleftrightarrow{k_{\pm 3}} NO + H \tag{8}$$

Where k_+ and k_- are reaction forward and backward Constants.

Assuming that consumption rate of free nitrogen atoms is equal to its production rate; *NO* concentration is obtained from following equation:

$$\frac{d[NO]}{dt} = 2k_{+1}[O][N_2]\frac{\left(1-\frac{k_{-1}k_{-2}[NO]^2}{k_{+1}[N_2]k_{+2}[O_2]}\right)}{\left(1+\frac{k_{-1}[NO]}{k_{+2}[O_2]+k_{+3}[OH]}\right)} \tag{9}$$

Constants of value of equilibrium reaction rate in the above equations are obtained in the reference [11]. Considering that the formation rate of *NO* is much lower than the original hydrocarbon oxidation rates, more thermal *NO* is formed after completing combustion. In above equation, concentrations of N_2 and O_2 are determined by combustion calculation and the radical concentration for [*O*] and [*OH*] is obtained from the following relations [12].

$$[O] = 36.64T^{1/2}[O_2]^{1/2}\exp\left(\frac{-27123}{T}\right) \tag{10}$$

$$[OH] = 212.9T^{-0.57}\exp\left(\frac{-4595}{T}\right)[O]^{1/2}[H_2O]^{1/2} \tag{11}$$

Therefore some source term in the part of the equation (5) is calculated from Zeldovich mechanism as follows:

$$S_{NO,th} = M_{NO}\frac{d[NO]}{dt} \tag{12}$$

In which M_{NO} is molecular mass of *NO* gas.

NO production fuel is a very complex phenomenon and it is strongly dependent to the flame stoichiometric, local combustion characteristics and initial concentration of the nitrogen compounds. Because of heating and evaporation of fuel droplets and nitrogen-containing radicals such as *HCN*, *CN* and *NH*, nitrogen-containing compounds are decomposed, which can be converted to NO_x. Considering that nitrogen cyanide (*HCN*) is predominant radicals, accepted mechanism for the formation of fuel *NO* is include of the formation of *HCN* from nitrogen in the fuel, and then performance of two oxidation reactions to get *NO* and combination with some part of *NO* and formation of N_2[13].

$$HCN \xrightarrow{+O_2} NO \tag{13}$$

$$HCN \xrightarrow{+NO} N_2 \tag{14}$$

Reaction rates of these two related based on measurements in De Soete work [13] are expressed as follows:

$$R_1 = A_1 X_{HCN} X_{O2}^{a}\exp\left(\frac{-E_1}{RT}\right) \tag{15}$$

$$R_2 = A_2 X_{HCN} X_{NO}\exp\left(\frac{-E_2}{RT}\right) \tag{16}$$

In the above equations, X is mole fraction and $A_1 = 3.5\times10^{10} 1/s$, $A_2 = 3\times10^{12}$, $E_1 = 67000 cal/mol$ and $E_2 = 60000$.

Considering that *NO* is produced in the reaction (13), and is used in reaction (14), source term from *NO* fuel in equation (5) is obtained from the following equation:

$$S_{NO} = (R_1 - R_2)\frac{M_{NO}}{RT}. \quad (17)$$

Numerical calculation

Dimension of computational mesh, is very effective on accuracy of results in the numerical calculations and on performance time. Although by increasing the number of nodes in mesh generation and computational field, the accuracy of results is increased, but much more time and memory are needed. In this study, the area near the entrance and walls that the flow properties changes are higher, smaller meshes are selected. Computing meshes in the combustion chamber is shown in Figure 2. Results have shown that applying the smaller meshes than 150 * 400 cells cannot make results better and this matter proved the mesh-independent solution. for discretization of convection terms in the governing equations, first order upwind scheme, and to correct the pressure field SIMPLE algorithm are used. For convergence, under relaxation factor of 0.7 for the flow field and 0.9 for the chemical species including pollutants are used. The convergence criterion of equations is $1*10^{-6}$. Numerical solution method is based on finite volume and steady state condition.

Boundary conditions

Due to axisymmetric flow, half of the combustion chamber in cylindrical coordinates in the form of two-dimensional is solved. Nozzle inlet diameter of liquid fuel, 1 mm and diameter of the fuel droplets, 50 to 100 microns are assumed. Fuel and air input temperature, 300K and inlet pressure, 1 *atm* has been considered. Central air inlet velocity is 2.5 m/s and the equivalence ratio is 0.66. Furnace wall temperature is assumed constant and equals to 750 K.

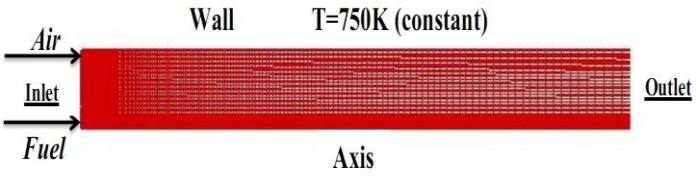


Figure 2. Computational mesh in solution domain.

Results and discussion

The flow streamline pattern inside the furnace, for the present numerical solution is shown in Figure 3. As shown in Figure 3, the vortex flow and backflow are informed near the furnace wall and entrance expansion area.

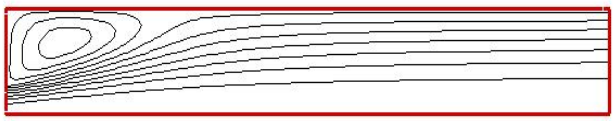
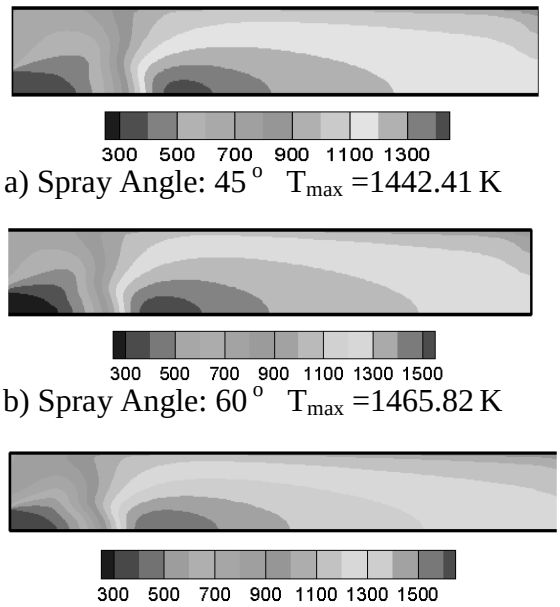


Figure 3. Stream lines inside the combustion chamber.

Temperature contours in the combustion chamber for different fuel spray angles of 45^o, 60^o and 80^o degree is shown in Figure 4. Increasing the fuel spray angle, the maximum temperature is increased. At spray angle of 45^o, 60^o, 80^o degree, maximum temperature are increased up to 1442 K, 1466 K, 1592 K respectively.



a) Spray Angle: 45^o T_{max} =1442.41 K

b) Spray Angle: 60^o T_{max} =1465.82 K

c) Spray Angle: 80^o T_{max} =1591.82 K

Figure 4. Temperature contours in combustion chamber for different fuel angles.

In Figure 5 the effect of the fuel spray angle on the temperature at the central axial of furnace is shown. Results show that with increasing the fuel spray angle, the amount of maximum temperature in the central axial furnace is also increased. By increasing the fuel spray angle, the time of staying particles inside the combustion chamber is increased. Due to extension of flame surface, axial velocity of particles is reduced and therefore the location of occurrence of maximum temperature in the central line of furnace with increasing the angle of fuel spray is closer to the input area.

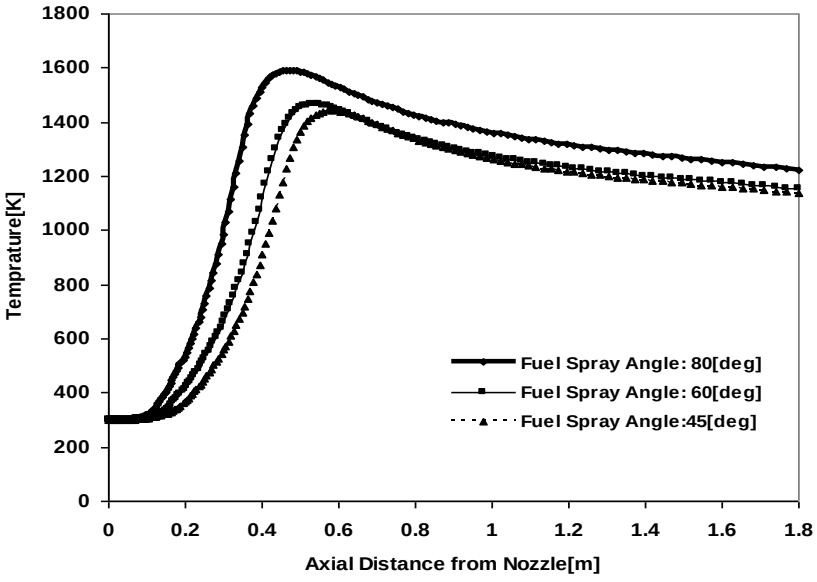


Figure 5. Effect of fuel angle on the temperature profiles on the central axial of furnace.

In Figure 6 NO_x Contour based on ppm has been shown for three fuel spray angle 45^o, 60^o and 80^o. It should be note that in the fluent software the output of NO_x concentration are given based on mass or mole ratio or mole concentration not in terms of ppm, which is common in the industry. An empirical function is defined and therefore the Fluent software can determine the amount of NO_x, in terms of ppm instead of mass or mole ratio.

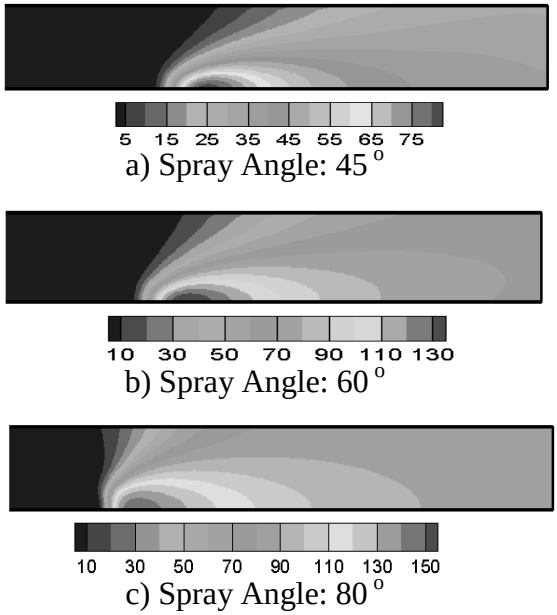


Figure 6. NO_x contour [ppm] at different fuel spray angles inside the combustion chamber.

The effect of spray angle on the NO_x emissions on the central axial furnace is shown in Figure 7. The results show that *NO* concentration along the axis is firstly increased from the burner to a maximum and then after slightly decreasing, remains constant almost to the end of the furnace. Three spraying angles comparison show that with increasing the angle of spraying, NO_x concentration is increased near the fuel nozzle. Fuel injection at low spray angles causes to longer flame length, thus the location of occurrence of maximum NO_x moves to farther distance in furnace from the input area.

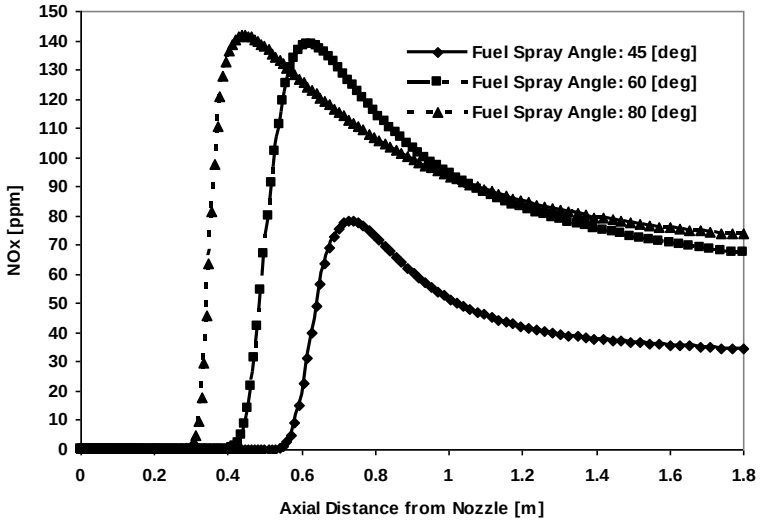


Figure 7. Effect of fuel spray angle on the NO_x concentration of in the central axial of furnace.

In Figure 8 the laboratory results for the effect of fuel spray angle on *NO* and NO_2 emissions in furnace output, with two different spray patterns as hollow and solid with equivalence ratio of 0.66 is shown. Increasing the spray angle causes to incensement of *NO* emission. Higher *NO* concentration is due to increasing the fuel retention time in the furnace and increasing of temperature because of its complete combustion, and thus creating suitable conditions for the formation of thermal *NO*.

As shown in Figure 8 with increasing the nozzle spray angle, NO_2 emissions in the furnace output is increased. However, NO_2 concentration compared with *NO* concentration is minimal, because approximately 90 to 95 percent of production of NO_x in combustion process is in form of NO. NO_x profile strongly depends on the flame temperature. By increasing angle of spraying, the fuel particles diameter in the output nozzle becomes smaller. This increases the contact surface between fuel and air and also the better mixing between them. Thus increasing the spray angle mines more complete combustion with higher maximum flame temperature [14].

The results show that pollutant emissions for the hollow cone spray pattern are approximately 10% less than solid cone. Nozzles with the hollow spray pattern are used for better fuel atomization, due to the more efficient radial distribution of fuel. In the hollow cone, the concentration of fuel droplets in the outer of spray edge is high, thus there is no fuel, even very low levels, in the center of spray. Solid cone spray pattern naturally produces a very long flame and it is not used in square or cylinder combustion chamber. Hollow cone, under inconsistent conditions, is more stable in spray angle and droplet distribution compared to solid cone. This matter for the fuel with high viscosity is remarkable, which may lead to reduce the effective spray angle and to increase the droplet size [15].

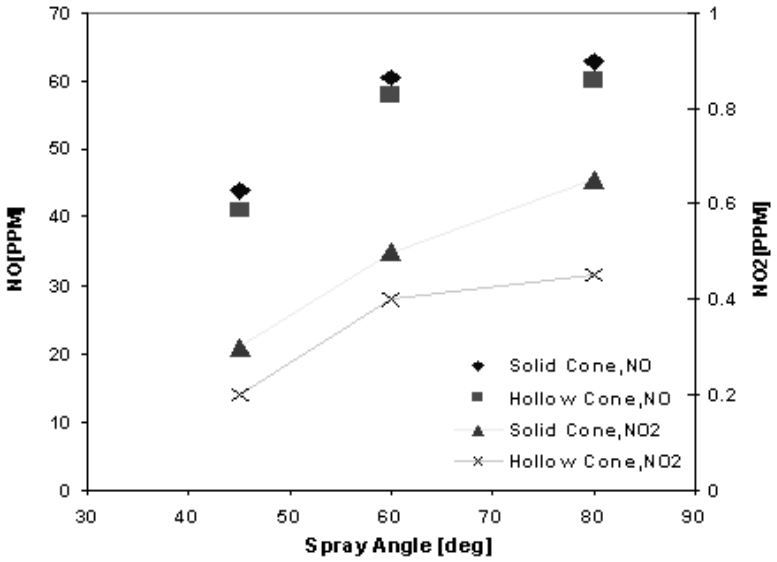


Figure 8. Effect of fuel spray angle on *NO* and NO_2 emissions in two different spray patterns (experimental results).

Effect of spray angle on NO_x emissions at furnace output in the form of laboratory and numerical computations are shown in Figure 9. Comparison of computational and experimental results shows that tendency of both methods for NO_x is similar, but computational results are slightly more than experimental results. High calculated temperature level is one of the reasons for this increasing. According to the thermal mechanism, high calculated temperature level is affected to the amount of simulated *NO*. This is more remarkable at the spray angle of 80^o because the calculated temperature compared with angle

of 45° is approximately increased 150K. Other reasons for reducing the amount of NO_x in the furnace output compared with numerical results is, reducing the temperature due to conduction heat transfer from the furnace wall, radiation heat transfer and back flow of cool gas in the end of furnaces, which is because of rotational combustion gases.

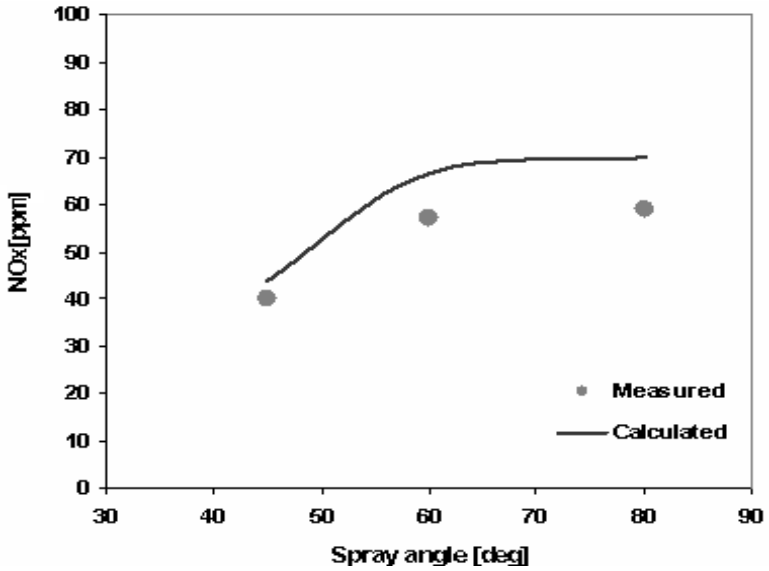


Figure 9. Effect of fuel spray angle on NO_x concentration in the furnace outlet, the comparison between numerical and experimental results.

Conclusion

Experimental and computational results for the pollutant emissions resulting from combustion of liquid fuels are evaluated. Results of laboratory measurements for three different fuel spray angles of 45°, 60° and 80° and two spray patterns are presented. Numerical simulations are done using fluent software version 6.32. Regarding to the existence of nitrogen in liquid fuels, the concentration of *NO* gas are calculated using two methods of thermal *NO* and fuel *NO* mechanisms. Comparison of computational results with experimental results shows good agreement. The results show that the amount of emissions is influenced of both spray angle and nozzle spray pattern. Increasing the fuel spray angle causes to increase NO_x in furnace output. Pollutant mission rate in solid cone spray pattern, at the equivalence ratio 0.66 is approximately 10 percent more than the hollow cone spray pattern. Numerical results also show that on the central axial furnace, NO_x gas concentration and temperature are in maximum value and by increasing the fuel spray angle, the maximum amount of temperature and NO_x in the central line are increased and move closer to the burner.

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