

Modeling of Pollutants Prediction from Fuel Burning in Oil and Gas Refineries

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ABSTRACT

Oxides of nitrogen ($\text{NO} + \text{NO}_2$, (NO_x)) are emitted from refineries, not only contribute to the production of photochemical smog at ground level but also cause damage to plant life and add to the problem of acid rain. The small amounts of prompt NO produced in the furnace chambers and oil refineries because the formation of NO_x can be attributed to four distinct chemical kinetic processes: thermal NO_x formation, prompt NO_x formation, fuel NO_x formation, and intermediate N_2O .

In this paper, the portions of pollutants that resulted in fuel burning (liquid or gas) in oil and gas refinery were studied by modeling of emitted gases in furnace chamber. The case study at Nassiriya power plant with different loads (70 – 210) MW was studied. The method of finite volume was studied to predict the pollutant portions by using FLUENT computer code (FLUENT is one of largest codes of computer programs which solve thousands of flow and combustion cases. The case study was drawn graphically then imported to solve by FLUENT). These types of pollution species are NO_x and SO_x as the important air pollutant influenced the human health. The numerical analysis in calculating the pollutants of chamber gave the findings of crude oil emission in combustion is higher than that at using gaseous fuel.

So, the methods of decreasing NO_x and SO_x pollution by water injection and exhaust gas recirculation are used in refinery operation were presented.

Keywords: Pollutant, Pollution, NO_x , SO_x , Combustion.

نمذجة التنبؤ بالملوثات من احتراق الوقود في مصافي النفط والغاز

الخلاصة

تنبعث أكاسيد النيتروجين ($\text{NO} + \text{NO}_2$) من المصافي، وتساهم ليس فقط في إنتاج الضباب الدخاني الكيميائي الضوئي على مستوى سطح الأرض ولكن أيضا تسبب ضررا على الحياة النباتية وتضاف إلى مشكلة الأمطار الحمضية. يعزى تكون كميات صغيرة من أكاسيد النيتروجين في غرف الفرن ومصافي النفط إلى أربعة عمليات حركية كيميائية منفصلة هي: تكون أكاسيد النيتروجين الحرارية، تكون أكاسيد النيتروجين الفورية، تكون أكاسيد النيتروجين من الوقود، و(N_2O) الوسطية. في هذا البحث، تمت دراسة مكونات الملوثات التي نتجت عن حرق الوقود السائل أو الغاز في مصفاة النفط والغاز والتي درست بنمذجة الغازات المنبعثة في غرفة الفرن. وقد درست حالة الدراسة في محطة توليد كهرباء الناصرية بأحمال مختلفة (70-210 MW). ودرست الحالة بطريقة الحجم المحدد للتنبؤ بنسب الملوثات باستخدام برنامج الكمبيوتر (FLUENT) والذي هو واحد من أكبر البرامج لحل الاف الحالات للجريان والاحتراق. اعتمدت دراسة الحالة بيانيا ثم استيرادها للحل بواسطة (FLUENT) هذه الأنواع من عناصر التلوث هي أكاسيد النيتروجين وأكاسيد الكبريت هي من اهم ملوثات الهواء التي تؤثر على صحة الإنسان. أعطى التحليل العددي في حساب الملوثات في غرفة الاحتراق نتائج لانبعاثات احتراق النفط الخام أعلى من ذلك في استخدام الوقود الغازي.

لذلك، قدمت الدراسة استخدام أساليب خفض أكاسيد النيتروجين وأكاسيد الكبريت الملوثة عن طريق حقن المياه وإعادة تدوير غاز العادم عند تشغيل المصافي.

كلمات مفتاحية: ملوثات، تلوث، أكاسيد النيتروجين، أكاسيد الكبريت، احتراق.

INTRODUCTION

Among chemical industries, petroleum refineries have been identified as large emitters of a wide variety of pollutants. Benzene, toluene, ethylbenzene, and xylene form an important group of aromatic volatile organic compounds (VOCs) because of their role in the troposphere chemistry and the risk posed to human health. Baltrenas et. al. (2011) showed that the concentration of benzene, as strictly regulated air pollutant by EU Directive 2008/50/EC, did not exceed the limit of 5 ppbv in the region in the vicinity of the crude oil refinery during the investigated period. No significant change in air quality in the vicinity of the oil refinery was discovered, however, an impact of the industry on the background air quality was detected [1]. However, the industrial sector is still an important source of environmental pollutants and thus modern environmental technologies are applied there [2]. Among chemical industries, oil refineries have been identified as large emitters of a wide variety of pollutants [3]. Organic micro pollutants such as volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) or polychlorinated biphenyls (PCBs) have been found in ambient air near petrochemical industries [4 - 6]. VOCs, mainly hydrocarbons, originate from production processes, storage tanks, transport pipelines and waste areas [7 - 10]. Typical compounds from petrochemical industries are: methane, ethane, propane, benzene, toluene, and xylene

[11]. In addition, oil refineries are associated with the emission of sulphur compounds, nitrogen oxides and particulate matter [8 – 10, 12]. The crude oil refineries have been linked to an increase of environmental pollution, as well as some adverse health effects for workers and people living nearby. A link between the presence of oil refineries and a varied symptomatology, including respiratory problems, leukemia, birth defects and adverse pregnancy outcome (preterm delivery) in nearby areas has been reported elsewhere [13 - 15]. Short-term exposure to particular concentrations of some VOCs present in the air is not considered acutely harmful to human health; however, long-term exposure may result in mutagenic and carcinogenic effects. Classic neurological symptoms associated with VOCs are fatigue, headaches, dizziness, nausea, lethargy and depression [10, 16 - 19]. A United States study emphasized that ambient VOCs attribute to 35–55% of the outdoor air cancer risk in the United States [20]. Several VOCs (benzene, toluene, ethylbenzene, xylene, isoprene, i, l, n-pentene) are also precursors of ground-level ozone formation [21], and they are mainly emitted in areas of intense transport and industrial activity [22]. Today, European air quality standards set the maximum level of benzene, which is $5 \mu\text{g}\cdot\text{m}^{-3}$ since January 2010 [23]. Benzene, toluene, ethylbenzene, and xylene (BTEX) form an important group of aromatic VOCs because of their role in the troposphere chemistry and the risk posed to human health [24]. In urban areas, BTEX constitute up to 60% of non-methane VOCs [25]. In the process of long-range transport, BTEX are among other VOCs that react with other pollutants such as NO_x and produce secondary pollutants with different reaction rates. Highly reactive species react near the vicinity of the emission sources, while slow reacting ones may be transported to a longer distance from the sources [26]. Hannun predicted the parameters inside the combustion chamber of power chimney by using similar eight burners consist of vents for air and fuel without swirlers [27]. The objectives of this study are: (1) to estimate the portions of NO_x at burning the different types of fossil fuel (liquid and gas); (2) to discuss the method to reach high efficient combustion at oil refinery stations.

NUMERICAL MODEL

Description of furnace

In this study, a furnace of power plant boiler was depended because it receive both oil and gaseous fuel for burning (Nasiriya power plant, Iraq). The shape of the furnace chamber is prismatic; the dimensions are 7.68×18. m. Furnace chamber capacity is 2160 m³ as shown in Figure (1). Rear wall of the upper part of furnace forms aerodynamic project of 2500mm depth. Furnace chamber is provided with 12 turbulent gas - oil burners, mounted on the rear wall in two stages. The burners of the lower stage are provided at the point 6150mm, the burners of the upper stage at the point 8750mm from the furnace bottom. Burners are provided for the separate gas and fuel oil burning systems as shown in Figure (2).

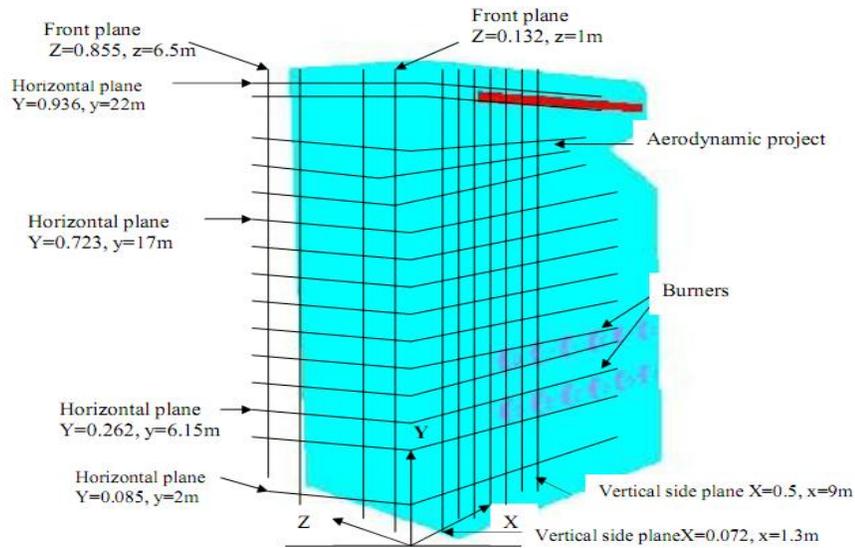


Figure (1) Furnace chamber.

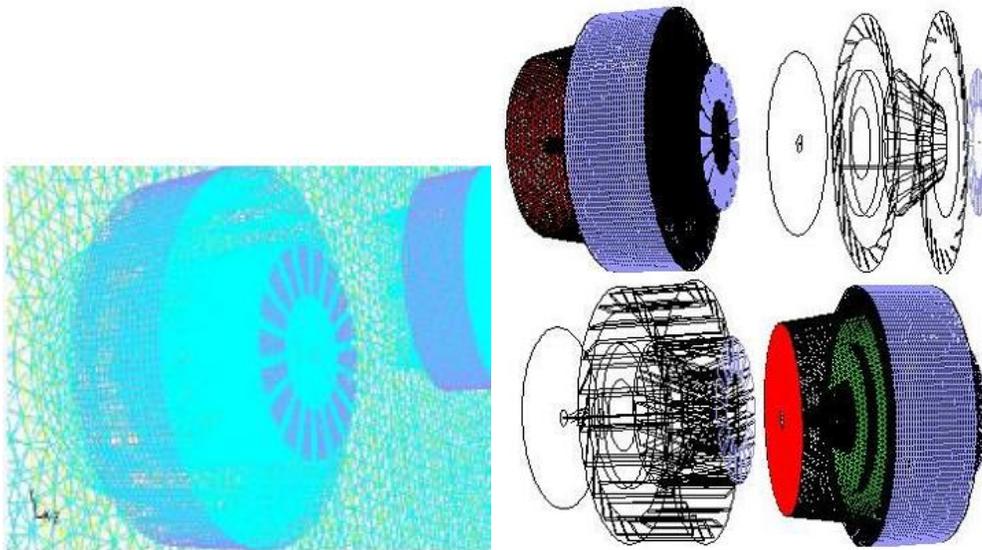


Figure (2) Burner design.

MATHEMATICAL MODEL

The gaseous fuel was natural gas with high ratio of methane which burns according to the following chemical reaction (complete combustion):



The molecular weight of carbon (C), hydrogen (H) and oxygen (O) are 12, 2 and 32 respectively. Therefore, the combustion of one mole of methane with two moles of oxygen produce one mole of carbon dioxide with two moles of water, that means one kg of methane with four kg of oxygen to produce the following:

$$M_{\text{CO}_2} = m_{\text{CH}_4} \times \frac{W_{\text{CO}_2}}{W_{\text{CH}_4}} \quad \dots (2)$$

$$M_{\text{H}_2\text{O}} = m_{\text{CH}_4} \times \frac{W_{\text{H}_2\text{O}}}{W_{\text{CH}_4}} \quad \dots (3)$$

The combination of the combustible fuel elements and compounds in the fuel with all the oxygen requires high temperatures enough to ignite the constituents, mixing or turbulence to provide intimate oxygen-fuel contact, and sufficient time to complete the process. Liquid fuel (crude oil) analysis via the south oil Refinery Company is given by Table (1).

Table (1) Combustion process analysis.

Products per kg of oxidant	Theoretical O2 required per kg of fuel	Reaction	Concentration by weight %	Fuel constituent	No
$\text{CO}_2 = \text{C}\% \times \frac{M_{\text{CO}_2}}{M_{\text{C}}}$	$\text{C}\% \times \frac{M_{\text{O}_2}}{M_{\text{C}}}$	$\text{C} + \text{O}_2 \longrightarrow \text{CO}_2$	0.865	C	1
$\text{H}_2\text{O} = \text{H}\% \times \frac{M_{\text{H}_2\text{O}}}{2M_{\text{H}}}$	$\text{H}\% \times \frac{M_{\text{O}_2}}{M_{\text{H}_2}}$	$2\text{H}_2 + \text{O}_2 \longrightarrow \text{H}_2\text{O}$	0.11199	H	2
$\text{SO}_2 = \text{S}\% \times \frac{M_{\text{SO}_2}}{M_{\text{S}}}$	$\text{S}\% \times \frac{M_{\text{O}_2}}{M_{\text{S}}}$	$\text{S} + \text{O}_2 \longrightarrow \text{SO}_2$	0.023	S	3

Specific gravity at 15.6 °C = 0.8641
 Where M_{O_2} , M_C , M_{H_2} , M_S , M_{CO_2} , M_{H_2O} , and M_{SO_2} are the molecular weight of oxygen, carbon, Hydrogen, sulfur, carbon dioxide, water and sulfur dioxide, neglecting the ash value because it does not enter the reaction process, i.e. from the above table:-
 The total mass of oxygen required per kg of fuel:

$$(m_{ox}) = C\% \times \frac{M_{O_2}}{M_C} + H\% \times \frac{M_{O_2}}{M_{H_2}} + S\% \times \frac{M_{O_2}}{M_S} \quad \dots (4)$$

But the concentration of oxygen in air is 23.36 by weight, then;

The exact weight of air required for combustion

$$\text{per kg of the fuel} = \frac{m_{ox}}{0.2314} \quad \dots (5)$$

Most gases involved in combustion calculations can be approximated as ideal gases, the reacting substances can be modeled by the equation of state [28].

$$r_{mix} = \frac{p/\bar{R}T}{\left(\frac{m_f}{M_f} + \frac{m_{ox}}{M_{ox}} + \frac{m_{pr}}{M_{pr}} \right)} \quad \dots (6)$$

Where:

$$m_{pr} = m_{CO_2} + m_{H_2O} + m_{SO_2} \quad \dots (7)$$

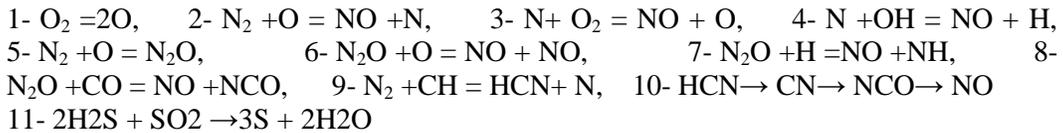
$$r_{CO_2} = \frac{p_{CO_2}/\bar{R}T}{m_{CO_2}/M_{CO_2}} \quad \dots (8)$$

$$r_{H_2O} = \frac{p_{H_2O}/\bar{R}T}{m_{H_2O}/M_{H_2O}} \quad \dots (9)$$

$$r_{SO_2} = \frac{p_{SO_2}/\bar{R}T}{m_{SO_2}/M_{SO_2}} \quad \dots (10)$$

Most of the nitric oxide (NO) formed in combustion consequently oxides to NO₂. For this reason it is customary to lump NO and NO₂ together and express results in terms of oxides of nitrogen (NO_x), rather than NO. It can be produced by four different

mechanisms: thermal NO, nitrous oxide mechanism, prompt NO and fuel NO [29]. The following reactions are possible to occur at the refinery chamber:



RESULTS AND DISCUSSIONS

The computational fluid dynamics (CFD) code consists of the following three main features:

1-Pre-processor: The task for the pre-processor is to gather the essential information needed for the solver to tackle the problem, i.e., the input. This involves: Defining the geometry of interest, i.e., the computational domain, generation of the grid, specifications of the physical and chemical phenomena that need to be modeled, definition of the fluid properties, and specification of appropriate boundary conditions.

2-Solver: The aim of the solver is to carry out the numerical calculations necessary to produce satisfactory simulations of the flow problem. Fluent uses a technique called the finite volume method (FVM). The numerical algorithm consists of the following structure:

- i. Integration of the governing equations of fluid flow over all the control volumes within the solution domain.
- ii. Discretisation involving substitution of a variety of finite difference-type approximations for the terms in the integrated equation representing the flow processes. The terms in equation are convection, diffusion and source terms. By doing this the system of integral equations are transformed into a system of algebraic equations that can be solved numerically.
- iii. Solution of the algebraic equations by an iterative method.

3-Post-processor: The post-processor returns the results of the simulation calculated by the solver. Today, most of the available CFD programs have developed graphical tools, which make it possible to receive a visualization of the calculated data.

The prediction of gases movement and aerodynamics inside the furnace and all other thermal, concentrations of species and products of combustion are studied. This work is carried out by using liquid fuel (Crude oil) and gaseous fuel (methane and mixture of contents) at different power loads of (210, 140, 70)MW.

Different iterations of NOx pollutants at non-premixed model with load (210MW) at no recirculation of flue gases are done. That for the total NOx pollutants (thermal and prompt), for NOx prompt pollutants and for NOx thermal pollutants, all them reach lower residuals to be stopped at convergence limit by using FLUENT code.

The total pollutant (NOx) for combustion of crude oil at load 210 MW, with no flue gases recirculation as vertical side planes is described by Figure (3). The maximum value is 0.0033kg/m³ at symmetric plane (X=0.5) closed to front wall (which

is high value in comparison with Table (2) and Figure (11). The other planes indicate high portions at the higher temperature situations. Therefore; the planes $X=0.072$ and $X=0.144$ have high pollutants. Null value is recorded at burners' entrance before the reaction and at the start of combustion.

Figure (4) demonstrates the pollutants NO_x for gaseous fuel furnace, the maximum value is 0.000871kg/m^3 at $X=0.216$ plane at the centre of furnace (which is lie at the same range of pollutants in Table (2) and Figure(11, 12). The total amount of pollutants here is lower than that in crude oil furnace because the combustion of gaseous fuel is rapidly occurring than in crude oil and the gas has few constituents (low carbon species).

The contours of prompt NO_x pollutants for crude oil are shown in Figure (5). It is the same as in Figure (3). The maximum value is 0.000017kg/m^3 at the symmetric plane ($X=0.5$), so, it is similar to Figure (6) with maximum value of 0.00328kg/m^3 because the total pollutants are the summation of prompt and thermal pollutants where this value lies among the values of Figure (12) that give the validity of this study.

The same as in Figure (4) for total pollutants of gaseous fuel; Figure (7) shows the same prediction for thermal NO_x with maximum value of 0.00087kg/m^3 and small amount of prompt NO_x didn't show here because it is gaseous fuel.

The NO_x pollutants amount for gaseous fuel furnace with different loads are demonstrated in Figure (8). The maximum NO_x pollution portions is at unit load 70 MW gradually decreased with increasing the load to minimum value at maximum load (210 MW). This is the reason of operating the flue gases recirculation ducts input to furnace at load 70MW while stop it at load 200-210MW, there are differences between the operation of flue gases recirculation which lessens the NO_x pollution amount.

Figure (9) indicates the summary of NO_x pollutants for different unit loads with crude oil fuel for two cases of operation (with recirculation of flue gases and without it), the curves give decreasing the pollutants with operation of flue gases except at loads more than 200MW which operates approximately at constant values of NO_x emissions.

The constitutes of gaseous fuel didn't contain species of sulphur, therefore the particulate of exhaust gas didn't include sulphur oxides or compositions (as shown in Table (2)). The portions of SO_x pollution as oil combustion in furnace are demonstrated in Figure (10) for all sections inside the chamber. These sections present a valid prediction approach to the estimated values of SO_x pollution in Table (2) because the crude oil spilled at south oil wells in Iraq has high portion of sulphur compositions.

CONCLUSIONS

According to the results of this paper, the following may be concluded:

1. The maximum NO_x emissions are at a load of 70MW gradually decreasing with increasing the load to minimum value at maximum load (210MW), this is the reason of operating the flue gases recirculation at load 70MW whilst stopping it at load of 200-210MW of power plant chamber.

2. Maximum NOx for crude oil fuel is concentrated at the line of symmetry of the furnace (X=9m) close to the back wall of furnace, because of the high temperatures gradient and low velocities, this is the reason for operating the flue gases recirculation at this position.
3. The pollution portions lie at the allowable levels of world fuel, with high NOx, and SOx particulate because of increasing the nitric oxides and sulphur oxides in the Iraqi oil.
4. To decrease the pollution of oil refineries, the operation conditions must be governed by lowering the temperature of chambers with method of flue gas recirculation and using the steam atomization spilled with fuel injection. The other method is by improving the fuel conditions at adding additives to ensure null pollution species before combustion. $2\text{H}_2\text{S} + \text{SO}_2 \rightarrow 3\text{S} + 2\text{H}_2\text{O}$

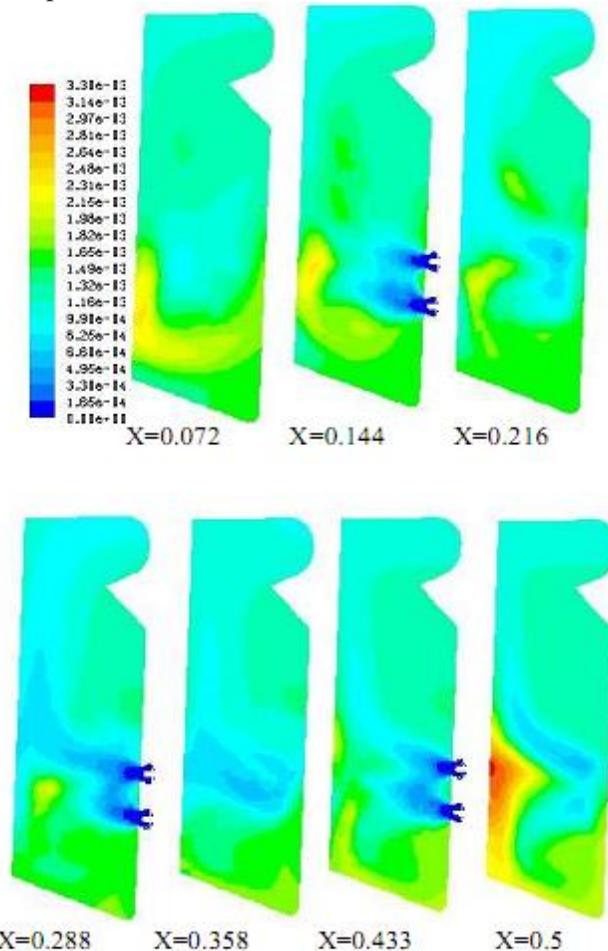


Figure (3) Contours of total pollutant of NOx (crude oil) at different side planes.

Table (2) Allowable Emissions, 10⁶ ton* [30]

Source category	TSP	SO _x	NO _x	CO
Electric utilities				
Coal	2.6	17.5	5.6	
Oil	0.2	1.9	1.4	
Natural gas	<0.1	0	0.9	
Total	2.8	19.4	7.9	0.3†
Percentage of grand total	20	65	31	0.3†
Other sources	11.0	10.4	17.8	112.2†
Grand total	13.8	29.8	25.7	112.5

*To obtain 10⁶ megagrams, multiply by 0.907.

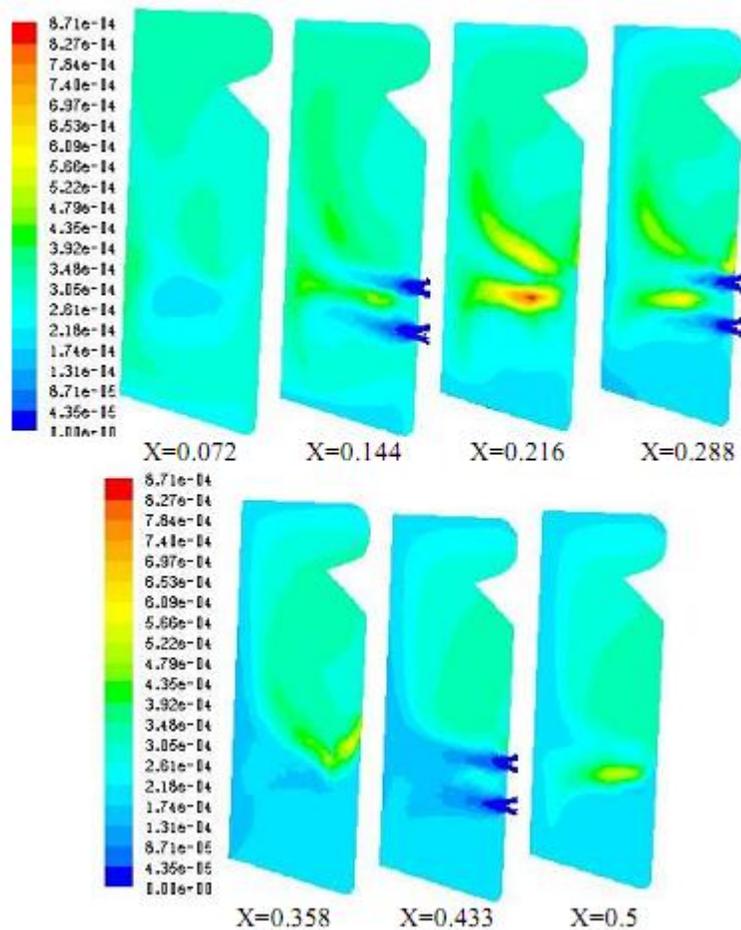


Figure (4) Contours of total pollutant of NO_x (gaseous fuel) at different side.

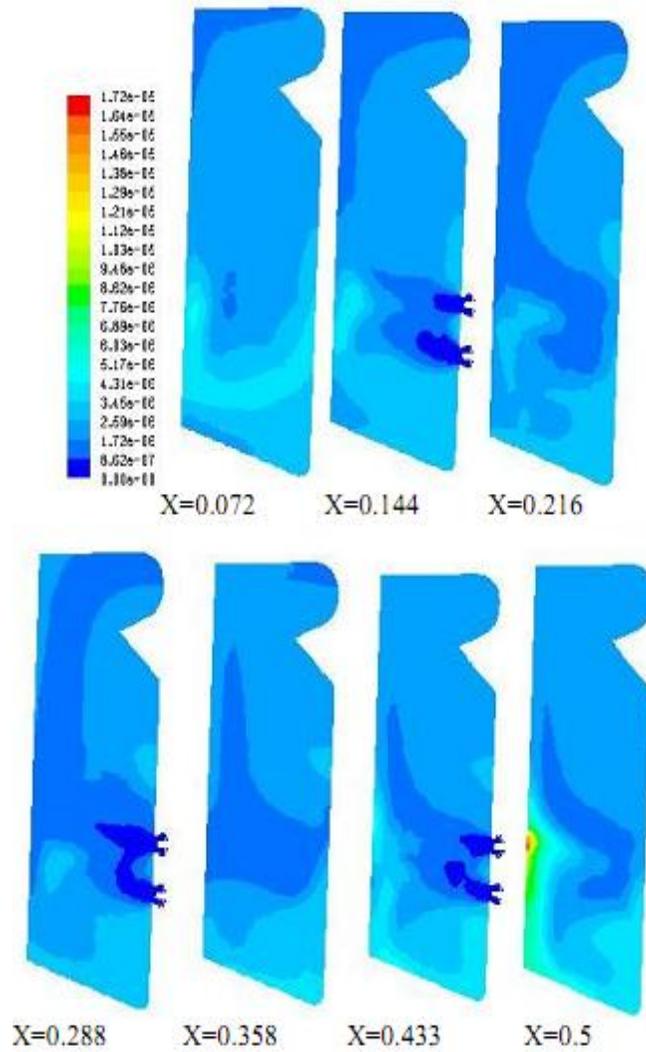


Figure (5) Contours of prompt NOx (crude oil) at different side planes.

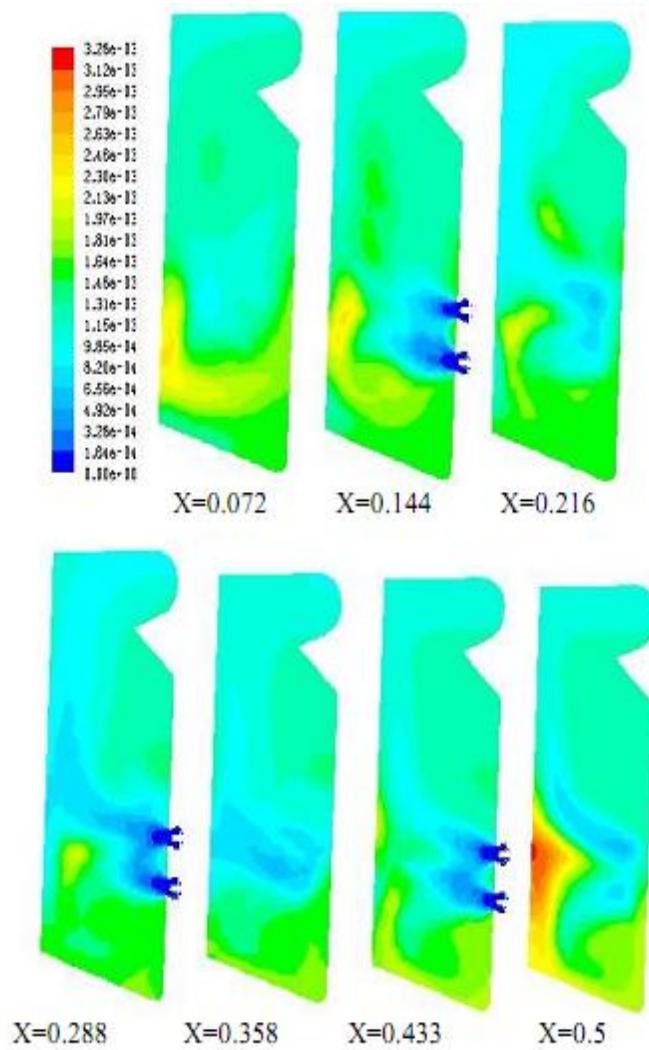


Figure (6) Contours of thermal NOx (crude oil) at different side planes.

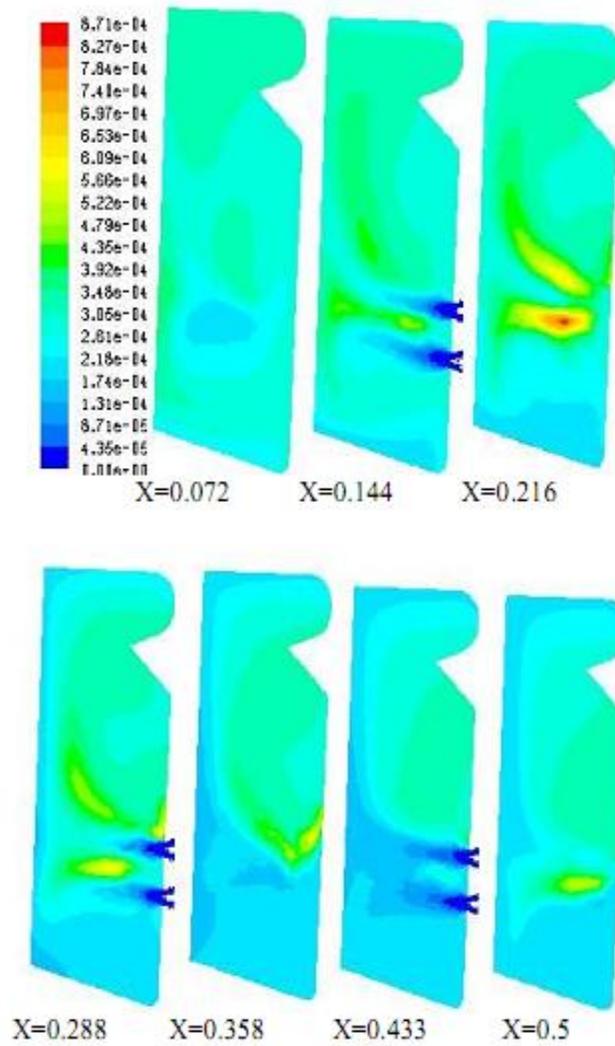


Figure (7) Contours of thermal NOx (gaseous fuel) at different side planes.

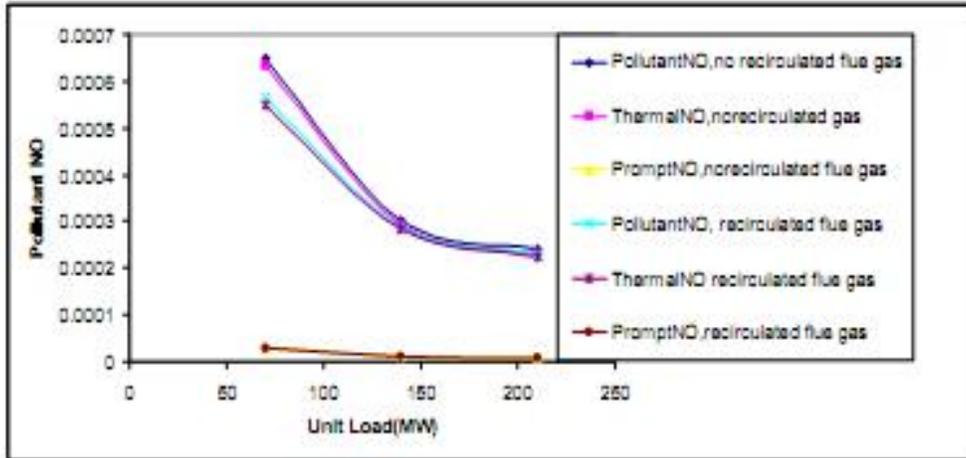


Figure (8) Pollutant NO for gaseous fuel with different load.

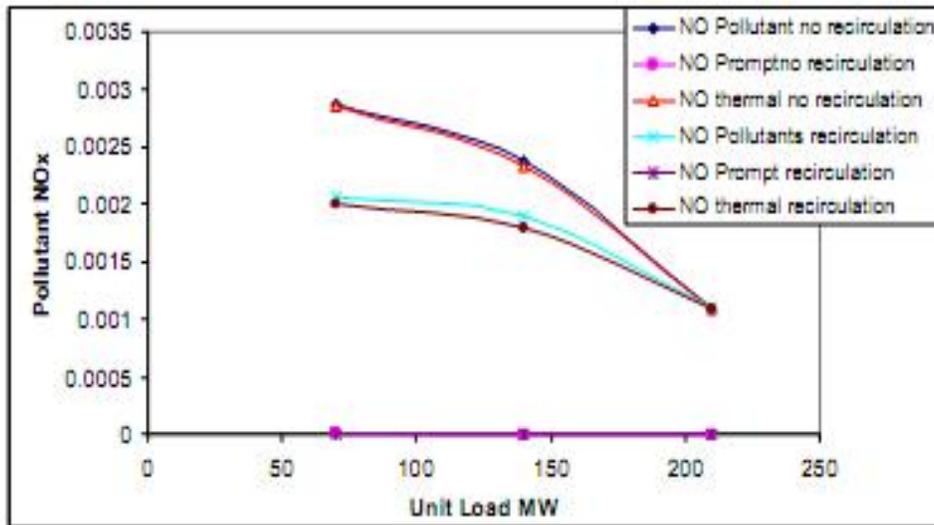


Figure (9) Pollutant NO for crude oil with different load.

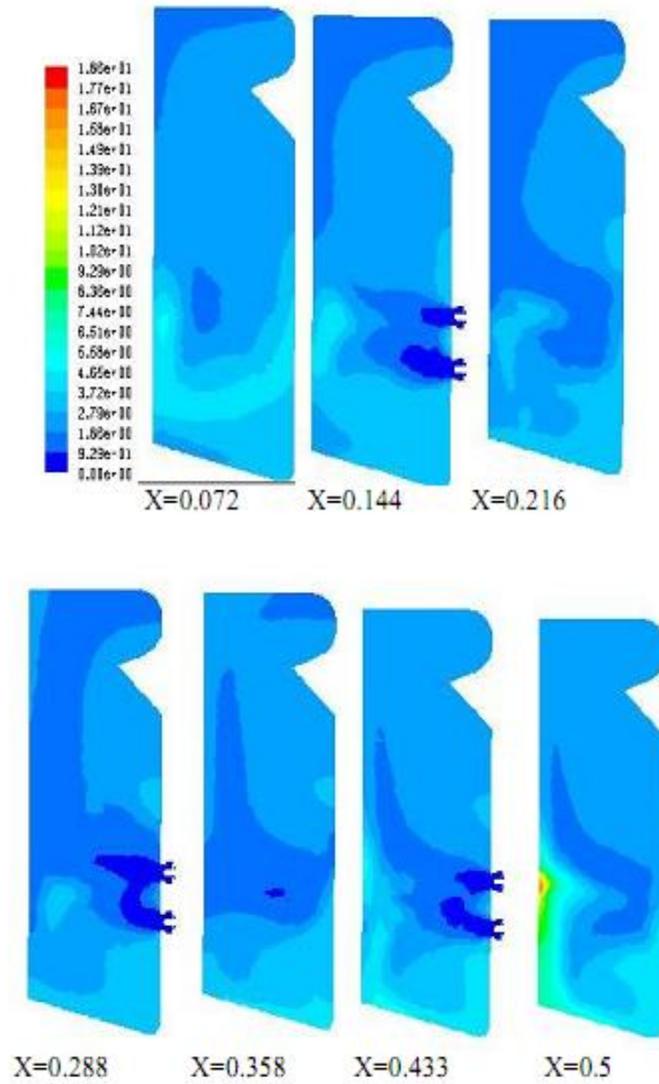


Figure (10) Contours of SOx for crude oil at different side planes

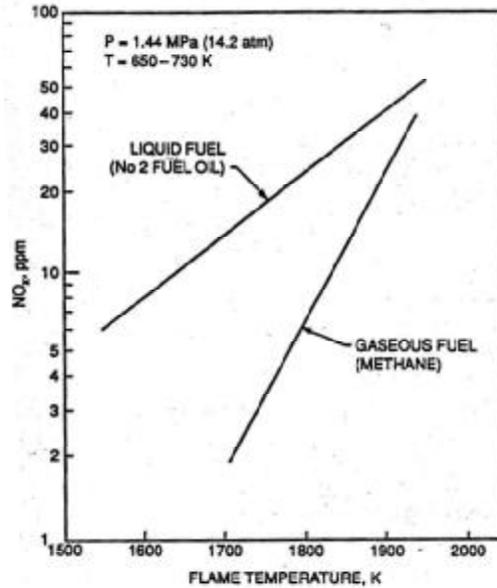


Figure (11) Dependence of NOx on flame temperature for liquid and gaseous fuel [31].

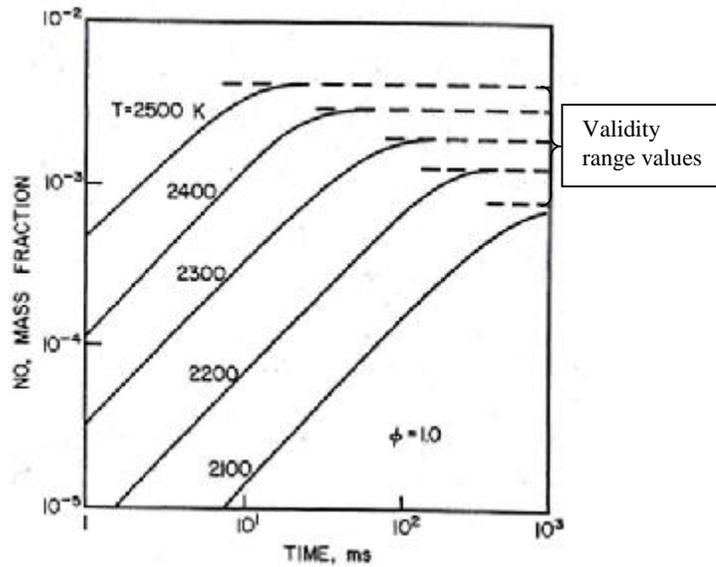


Figure (12) NOx formation as a function of time and temperature, P=1MPa [29].

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NOMENCLATURE

Symbol	definition	Symbol	definition
P	Static pressure (pa)	M_f, M_{pr}	Molecular weights of fuel and products of combustion respectively
ρ	Density (kg/m^3)	m_{pr}	Mass of products in kg per kg of O_2 .
μ_{eff}	Effective viscosity ($N.s/m^2$)	\bar{R}	Universal gas constant in $kJ / kmol.K$
\dot{m}_f	Mass flow rate of fuel (kg/s)		
T	Temperature in Kelvin.		
m_a	Mass flowrate of the air (kg/s)		