Combustion Emissions and Thermal Performance Impacts When Replacing Hydrocarbon Fuels with Hydrogen in Industrial Furnaces

Marc Cremer and Dave Wang
Reaction Engineering International
cremer@reaction-eng.com

Yong Wang
TechnipEnergies
yong.wang@technipenergies.com

Abstract

Many industrial processes rely on heating that is currently achieved through combustion of fossil fuels. The industrial sector generates approximately 23 percent of the greenhouse gas emissions in the US. As infrastructure associated with production and distribution of hydrogen continues to expand, replacement of carbon containing fossil fuels with non-carbon hydrogen for process heating in industrial plants is becoming an interesting option for reductions of greenhouse gases in the industrial sector. Hydrogen has several characteristics that create challenges when considering replacement of natural gas or other hydrocarbon fuels. The high adiabatic flame temperature and high flame speed of hydrogen-air combustion can lead to significant increases in NOx emissions, as well as large impacts to the thermal profile. The high flame speed limits the practical ability to rely on lean pre-mixing to mitigate high NOx emissions, as has been successfully implemented in low NOx natural gas burners. However, the wide flammability limits of hydrogen make fuel and air staging, along with induced flue gas recirculation, attractive options for limiting NOx emissions.

Both 1-D process modeling and 3-D computational fluid dynamics (CFD) modeling are useful tools which will be heavily utilized for assessment of NOx emissions and thermal performance as industrial boilers and furnaces convert to higher concentrations of hydrogen in fuels. Simulations of idealized reactor networks with well verified detailed chemical kinetic mechanisms effectively show how fuel and air staging, as well as gas recirculation, can impact the peak flame temperature to reduce NOx emissions. These calculations can be used to approximate the degree of staging and/or the amounts of gas recirculation that must be achieved to target specific NOx emission limits. These estimates can be further refined by incorporation of realistic descriptions of fuel and air mixing, coupled with finite rate chemical kinetics within CFD simulations for assessment of NOx emissions and heat flux distributions. This paper will show results of both idealized reactor network calculations and 3-D CFD simulations to quantify impacts of hydrogen air combustion compared to natural gas combustion in a full-scale industrial furnace.

1 Introduction

The US EPA reported total CO_2 emissions in the US in 2019 to be 6,558 million metric tons [1]. Sources from industry and electricity production contributed to just under 50 percent of all emissions, as shown in Figure 1. Over the past decade, CO_2 emissions from the electricity sector have shown significant reductions as power generators have retired coal plants, replacing them with renewable sources or with higher efficiency gas turbines. In some cases, power generators have chosen to retain existing coal boiler assets and convert them to natural gas, thereby reducing CO_2 emissions due to the lower C:H ratio of natural gas compared to coal.

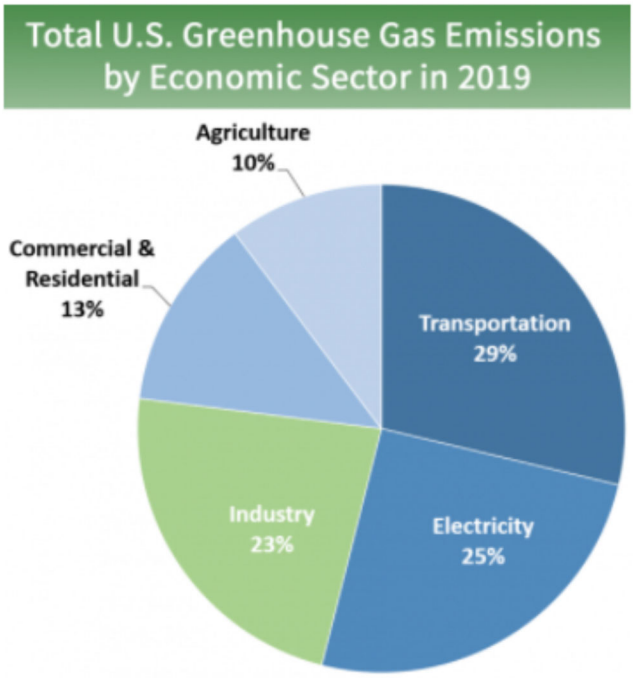

Figure 1. Fraction of US greenhouse gas emissions by sector in 2019 [1].

The industrial sector, however, has made less progress toward reduction of greenhouse gas emissions. A large fraction of industrial heating equipment is fired with natural gas or with refinery gases. Reducing or replacing consumption of these carbon containing fuel gases with hydrogen is an attractive option, particularly due to the lack of CO_2 and CO emissions. In addition, there are plans and progress toward development of infrastructure for production of large volumes of hydrogen. For example, the U.S. Department of Energy has developed an initiative called H2@Scale to bring together stakeholders to advance affordable hydrogen production [2]. Ten million metric tons of hydrogen are currently produced in the US every year, primarily through steam methane reforming. Deployments of clean alternatives, primarily electrolysis, are rapidly increasing.

Although hydrogen could be an effective strategy for decarbonizing the industrial sector, technical problems must be solved before widespread utilization of hydrogen will be achieved. Hydrogen has many characteristics that differ from those of natural gas, which impact both thermal performance and potential emissions. The primary product of combustion of hydrogen and air is water vapor. Since the adiabatic flame temperature of hydrogen is significantly higher than that of methane (e.g., 3807ºF vs. 3524ºF), there is potential for increased emissions of NOx. Per the extended Zeldovich mechanism, thermal NOx emissions become significant at flame temperatures above 2700ºF and increase exponentially at increasing temperature. As written by a directory of the National Centre for Atmospheric Science, increasing NOx emissions from industrial combustion equipment is not an acceptable trade-off for reducing CO_2 emissions, as NOx emissions are known to contribute to photochemical smog, which could disproportionately impact the urban poor.

In addition, hydrogen burns at a significantly higher flame speed as compared to natural gas (i.e., factor of 7). Existing ultra-low NOx natural gas burners successfully employ fuel lean premixing

and low swirl stabilization [3]. The high flame speed of hydrogen increases the potential for flashback if lean premixed low-NOx burner strategies are used [4]. Flashback is a serious safety issue which is exacerbated when the burner must be operated under turndown conditions where the reduced inlet velocities increase the potential for flashback.

To reduce NOx emissions from hydrogen-air burners, there has been significant research pertaining to high pressure lean-premixed flames for application in gas turbines, where the extremely high stoichiometric ratios (SRs) help to keep the flames cool, and mitigate thermal NOx emissions. In most industrial heating applications, these high SRs are not feasible due to poor efficiency. One very recent study utilized a common industrial gas burner but manipulated the number and size of the air nozzles to influence the air jet velocity and affect the entrainment of flue gases, and thereby reduce NOx emissions [5]. This study showed that the generation of NO_x from a hydrogen air flame can be more than 60 percent higher than for a natural gas flame in a commercial industrial gas diffusion burner due to the increase in adiabatic flame temperature. Additionally, this study also showed that by controlling fuel and air mixing to reduce the adiabatic flame temperature, NO_x emissions can be significantly reduced (i.e., less than a 10 percent increase over a natural gas flame).

In addition to the safety concerns of flashback, hydrogen's high flame temperature and high flame speed tend to produce short, high temperature flames which increase the likelihood of boiler and heater hotspots and a shift in heat balance towards the radiant section. So, it will not be sufficient to develop industrial burners that are low in NOx emissions. Burners will also need to be designed to closely match existing thermal heating profiles that are obtained with existing carbon-containing fuels, and developed over years of performance optimization efforts.

While the electricity industry has been quite successful in transitioning away from the use of coal in favor of natural gas as an approach to reduce their greenhouse gas emissions, US industry already utilizes a large fraction of natural gas and refinery gases in their heating processes. The US industry has identified that burning fuels that are high in hydrogen, up to 100 percent hydrogen, will require significant research and development to accommodate known differences in flame speed, adiabatic flame temperature, heating value, radiative properties, etc., which will significantly impact thermal performance and emissions. The impact of increasing H_2 utilization on CO_2 reduction is nonlinear, with the need for very high replacement rates of hydrocarbons with H_2 to achieve high percentage decreases in CO_2 emissions. It is also understood that existing burners of natural gas may not be flexible enough to burn gases exceeding 80 percent H_2. Thus, significant research efforts will be required by equipment users and designers to develop burner and firing configurations that concurrently limit NOx emissions and optimize thermal performance of existing industrial equipment burning fuels with high concentrations of H_2.

In this paper, we use chemical kinetic calculations and computational fluid dynamics (CFD) modeling to predict the impacts of increasing the fraction of H_2 in fuel blends with natural gas on both thermal performance and NOx emissions. The chemical kinetic evaluation examines the impacts of fuel staging and air staging as well as flue gas recirculation (FGR), on NOx emissions. The CFD evaluation investigates the impacts of replacing natural gas with 100 percent H_2 in two furnace configurations: 1) ethylene furnace, and 2) power boiler.

2 Methods

2.1 Description of CFD and Chemical Kinetics Models

Reaction Engineering International (REI)'s proprietary CFD code, *ADAPT*, was used for the evaluation reported in this paper. *ADAPT* is a three dimensional, reacting CFD code that has been extensively utilized to simulate gas-fired combustion applications requiring an accurate treatment of chemistry and turbulence-chemistry interactions [6, 7]. Finite rate chemical kinetics for the CFD simulations are implemented through use of reduced mechanisms developed from the Computer Assisted Reduced Method (CARM) [8], based on steady state assumptions. The detailed mechanism used to develop the reduced mechanism is the GRI3.0 mechanism (53 species and 325 reactions). The reduced mechanism used in this study contains 22 non-steady state species. In-situ Adaptive Tabulation (ISAT) is used to reduce the computational time associated with integration of the finite rate chemical kinetics. Turbulence-chemistry interactions are represented using the eddy dissipation concept (EDC).

The detailed chemical kinetics simulations are based on the detailed mechanism GRIMech3.0. Chemical kinetic networks are constructed using perfectly stirred reactor (PSR) and plug flow reactor (PFR) calculations in series or in parallel using REI's Reaction Engineering Kinetics Solver (REKS) package along with REI's Modlink graphical user interface package.

2.2 Description of Ethylene Furnace

The majority of current ethylene furnace designs involve a single or twin radiant firebox and a single convective section. Natural gas or refinery gas (with H_2 concentration up to ~50 percent by volume) are typically fired through floor burners, or a combination of floor and balcony or radiant wall burners. The coils are typically arranged in the center of the radiant box, being heated by the burners firing along the opposite walls. Burners typically use fuel staging and in-situ FGR to limit NOx emissions while maintaining the appropriate flame height and heat release profile to achieve the desired process heat flux profile, while avoiding flame rollover into the tubes.

For this study, a two-burner section of an ethylene furnace, representing a section or "slice" of the furnace with two floor burners firing along opposite walls, and a coil section in the center, was used to investigate the emission and thermal performance impacts when replacing natural gas with hydrogen. Two principals were held when replacing hydrocarbon fuel with hydrogen fuel: 1) maintaining the same SR (i.e., SR = 1.1 for this study); 2) maintaining the same fuel firing rate based on lower heating value (LHV). The floor burners are flat-flame burners with >85 percent fuel staging. For this study, the burner geometry was held fixed, including fuel orifice sizes, as the fuel content was changed.

Figure 2 shows the H_2 mole percentage in the fuel when the percentage of LHV-based firing rate of natural gas fuel is replaced by hydrogen fuel. Initial H_2 mole percentage in the fuel mixture raises quickly to 50 percent when the LHV firing rate replacement is ~ 24 percent. When the LHV firing rate replacement reaches 80 percent, the H_2 mole percentage in the fuel will be larger than 90 percent. Also noted in Figure 2 is the adiabatic temperature rise of 350°F for the H_2 fuel in comparison with natural gas fuel.

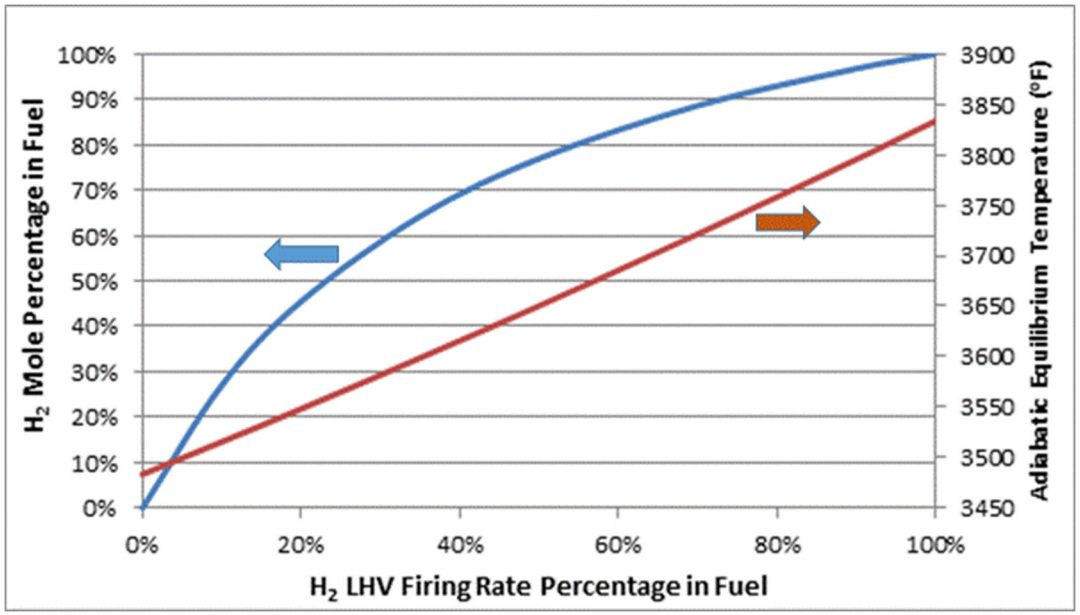

Figure 2: H_2 mole percentage in the fuel and adiabatic temperature vs LHV firing rate replacement percentage

2.3 Description of Power Boiler

For the evaluation carried out here, a front wall fired utility boiler configuration was chosen as shown in Figure 3. A dual register, low NOx burner of a generic design was assumed, as shown in Figure 3. The CFD simulations were performed with natural gas fuel and H_2 fuel. Two principals were held when replacing hydrocarbon fuel with hydrogen fuel: 1) maintaining the same SR (i.e., SR = 1.14 for this study); 2) maintaining the same fuel firing rate based on LHV.

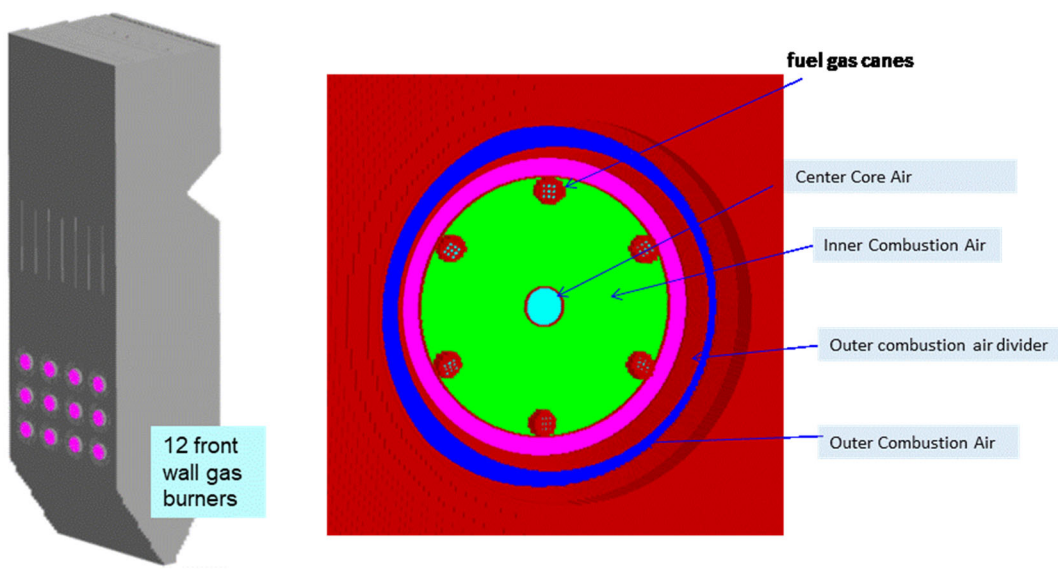

Figure 3: Power boiler and burner geometry

3 Results and Discussion

3.1 Chemical Kinetics Calculations

Figure 4 shows the results of calculated flame temperatures and NO_x concentrations with H_2-air combustion under the assumption of: 1) Thermodynamic equilibrium, and 2) Chemical kinetic rate limitations. Under the assumption of thermodynamic equilibrium, NO_x concentration depends only on the flame temperature and SR (i.e., oxygen availability). When the kinetic rate limitations are taken into account, the NO_x emissions are dependent on the time history associated with the local mixture composition and temperature. Different time histories with differing mixtures, residence times and temperatures can result in significant differences in NO_x emissions. The chemical kinetically limited calculations were based on a PSR, at a residence time of 0.4 sec and fixed atmospheric pressure. Figure 4 shows that for the chosen residence time, the chemical rate limited temperature matches the equilibrium temperature over the SR range. However, there is a large deviation between the equilibrium-based NO_x and the rate limited NO_x calculations, particularly at SRs > 1.0. The temperatures and NO_x concentrations calculated in Figure 4, due to the adiabatic condition, are much higher in reality than would typically be observed in an industrial furnace because of radiation heat loss and FGR.

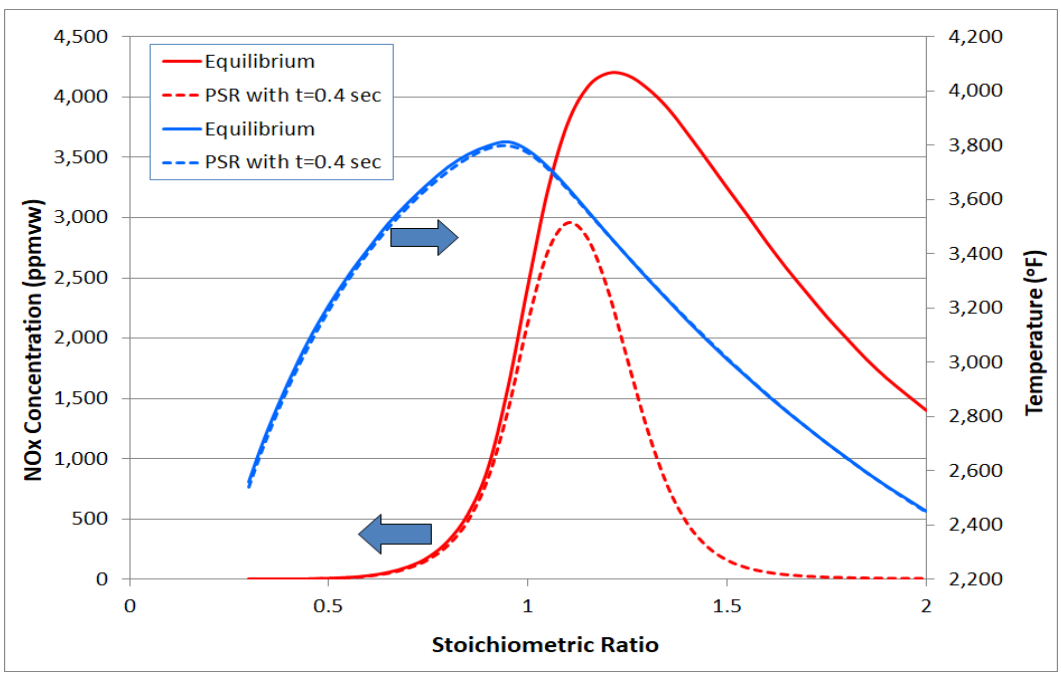

Figure 4: Calculated adiabatic temperature and NO_x concentrations in H_2-air flames based on equilibrium and finite-rate limited conditions

Figure 5 shows NO_x formation for natural gas-air, and H_2-air combustion at four fuel lean SRs. The calculations were based on a PFR with a residence time of 0.5 sec. The initial composition is assumed to be the mixture of combustion products along with 5 percent un-combusted fuel and excess air. Figure 5 also shows the significant NO_x formation (i.e., thermal NOx route) at high temperatures (> 2700°F) for both natural gas fuel and H_2 fuel. However, at the same SR, temperature, and residence time, H_2–air combustion produces about ~10 percent lower emissions

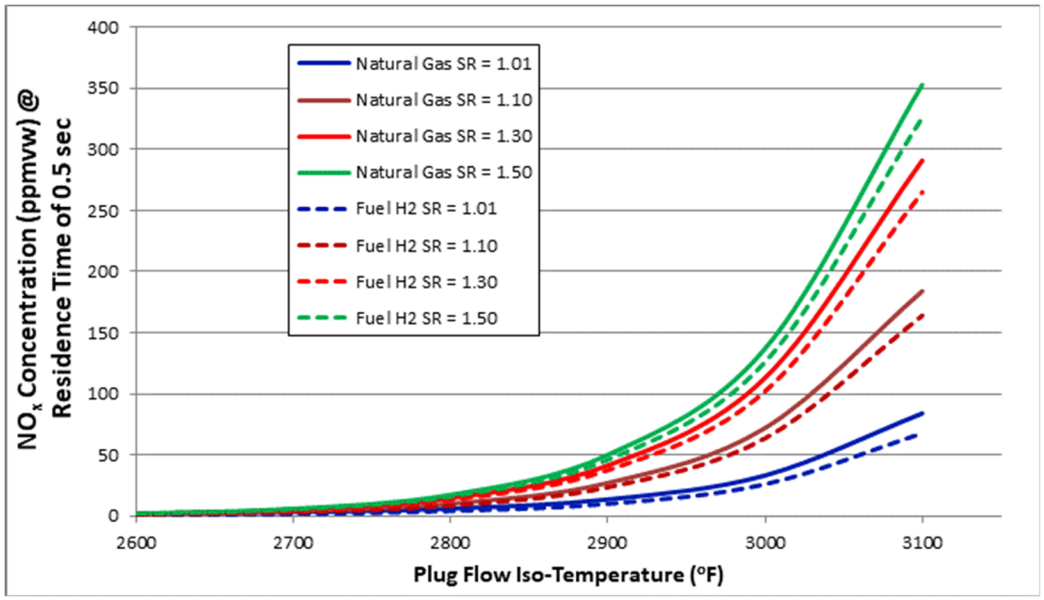

Figure 5: Iso-temperature thermal NOx formation for natural gas-air combustion and H_2-air combustion at four fuel lean SRs

(based on mole fraction, i.e., ppmvw) than that of natural gas–air combustion. Thermal NO_x is formed by the high-temperature reaction of nitrogen with oxygen via the well-known extended Zeldovich mechanism.

$$O + N_2 \leftrightarrow NO + N \quad \text{(Eq. 1)}$$

$$N + O_2 \leftrightarrow NO + O \quad \text{(Eq. 2)}$$

$$N + OH \leftrightarrow NO + H \quad \text{(Eq. 3)}$$

The H_2-air combustion produces more H radical than natural gas-air combustion, thus promoting the reverse Zeldovich reaction.

Figure 6 shows iso-temperature NO_x formation for natural gas-air combustion and H_2-air combustion at two fuel rich SRs. The calculations were based on a PFR with a residence time of 0.5 sec. The initial composition is assumed to be the mixture of completely combusted product with 5 percent un-reacted air and remaining excess fuel. 200 ppmvw NO is also added to the initial composition. Figure 6 shows that: 1) at high temperature (i.e., > 2700 ºF), H_2-air combustion is more effective in reducing NOx; 2) at low temperatures (< 2700ºF), NO_x is reduced mainly through the reburning mechanism and natural gas is more effective reburning agent at SR = 0.85.

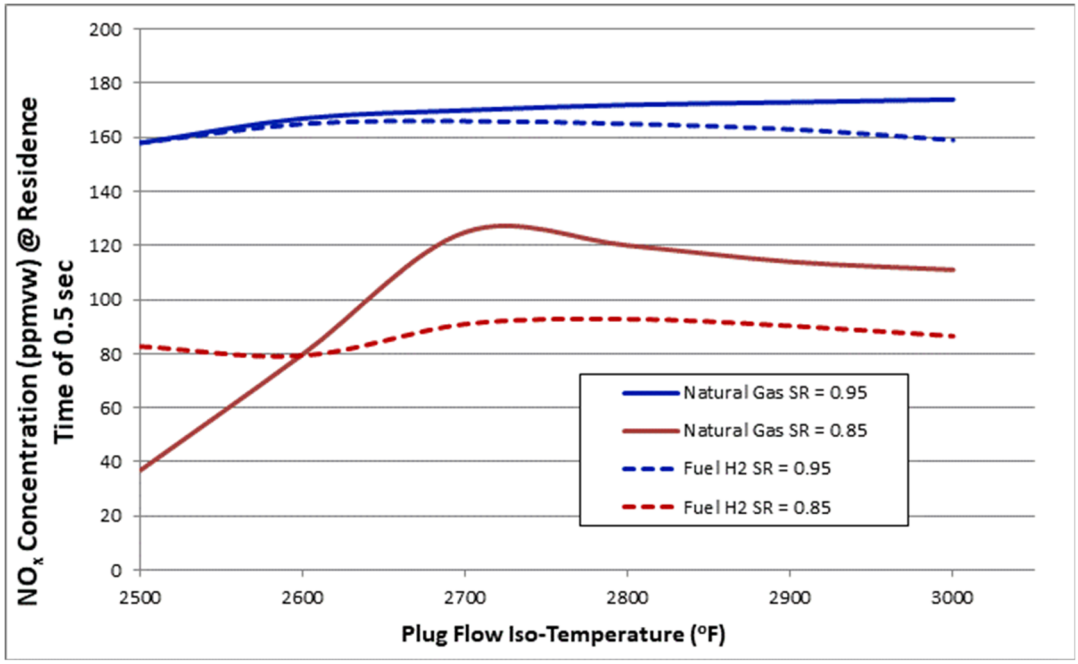

Figure 6: Iso-temperature NOx formation for natural gas-air combustion and H_2-air combustion at two fuel rich SRs with initial NO concentration of 200 ppmvw

A series of calculations were subsequently carried out using networks of PSRs and PFRs to represent NOx emissions associated with: 1) air staging, and 2) fuel staging. In both cases, varying quantities of FGR were assumed to be mixed into the flame. It was found that the final NO_x emission is highly dependent on the extent of staging, residence time, the amount of FGR, the heat loss associated with FGR etc. It is expected that NO_x emission and thermal performance behavior, when replacing hydrocarbon fuels with hydrogen fuel, will be highly dependent on the furnace type and operating conditions.

3.2 Results of Ethylene Furnace Simulations

Figure 7 shows the iso-thermal surface (2500°F and 2800°F) for natural gas fuel and H_2 fuel. These iso-surfaces are colored by the O_2 concentration. Natural gas fuel and H_2 fuel show the same shape for the iso-thermal surface of 2500°F, suggesting the overall flame shape is similar when replacing the natural gas fuel with H_2 fuel. The elevated temperature locally for the H_2 fuel is obviously shown in Figure 7 when comparing iso-thermal surface of 2800 °F for natural gas fuel and for H_2 fuel. However, due to fuel staging and relatively high rate of in-situ flue gas recirculation in this furnace, the level of local temperature increase is much less than the adiabatic temperature increase when replacing natural gas fuel with H_2 fuel. Local elevated temperatures result in increased radiation heat transfer to the coil, but does not necessarily mean high NO_x emission.

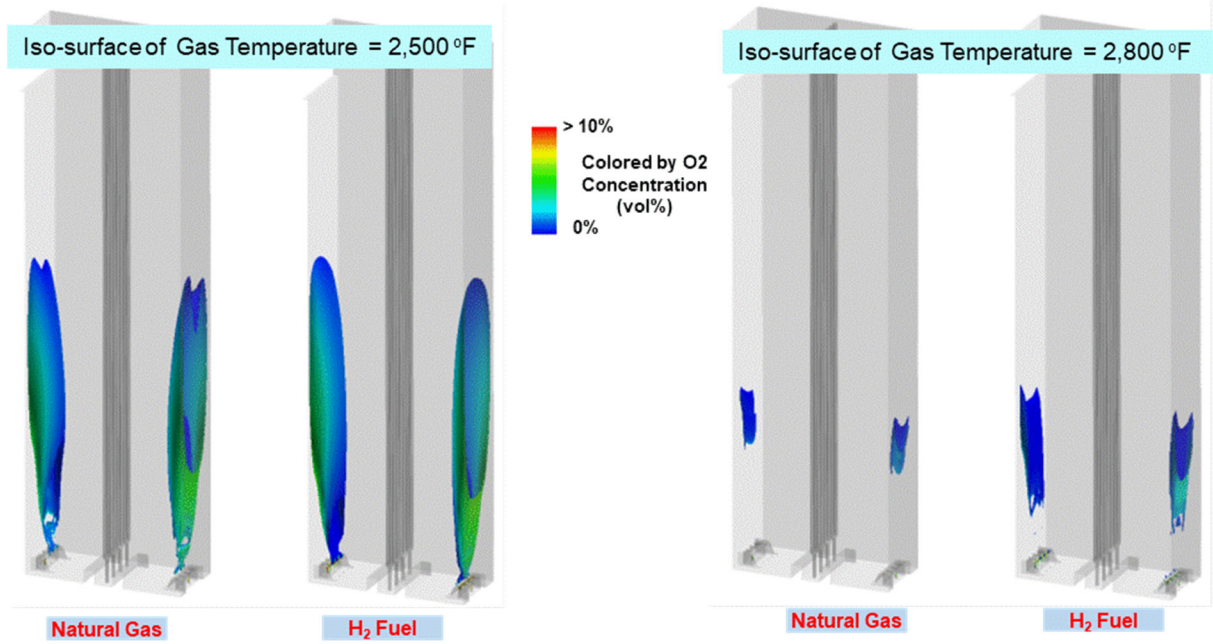

Figure 7: Iso-thermal surface of 2500°F and 2800°F when burning natural gas and H_2 fuel

Figure 8 shows the furnace radiant efficiency as a function of H_2 mole percentage in the fuel. As the H_2 mole percentage increases, the furnace radiant efficiency also increases. Furnace radiant efficiency increases by ~4.5 percent when natural gas fuel is completely replaced by H_2 fuel.

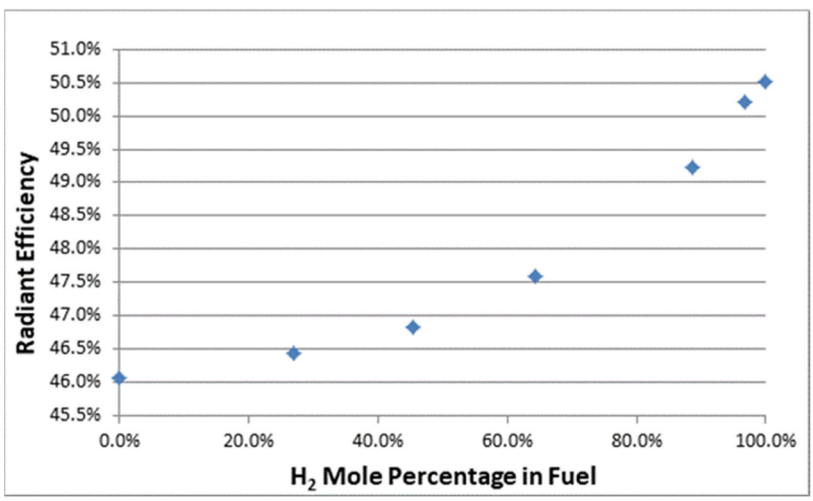

Figure 8: Furnace radiant efficiency as a function of H_2 mole percentage in the fuel

Figure 9 shows the NO_x emission expressed as ppm @ 3 vol% O_2 dry as a function of H_2 mole percentage in the fuel. NO_x emission expressed in other forms will be explored later in the power boiler section. As the H_2 mole percentage in the fuel increases, the NO_x emission increases initially. When the H_2 mole percentage in the fuel is larger than 50 percent, the NO_x emission starts to decrease as the H_2 mole percentage in the fuel increases.

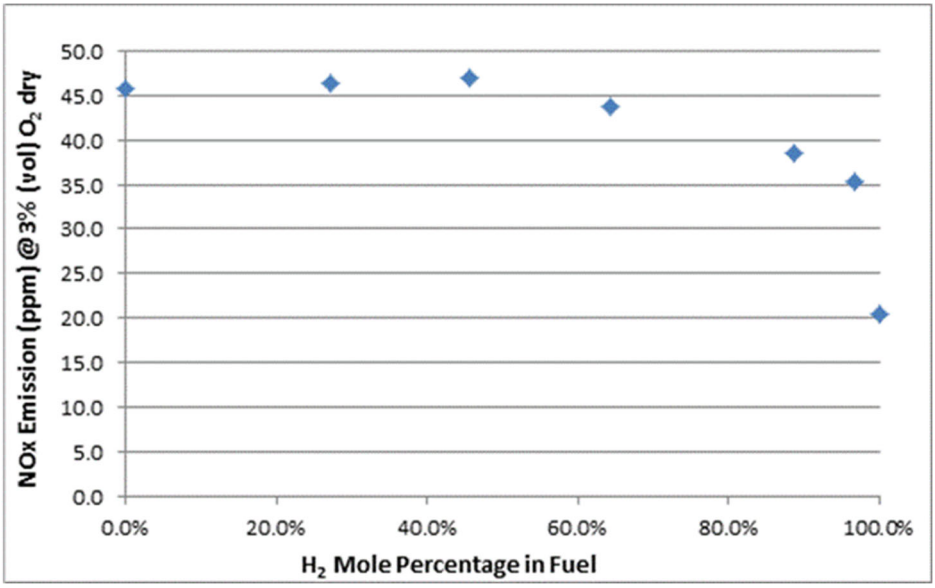

Figure 9: NO_x emissions as a function of H_2 mole percentage in the fuel

The NO_x emission decreases significantly as the H_2 mole percentage approaches 100 percent. This NO_x emission behavior is thought to be related to the influence of the in-situ flue gas recirculation in the ethylene furnace.

Figure 10 shows the reverse flow rate and reverse flow ratio vs. furnace height as a function of H_2 mole percentage in the fuel. The reverse flow rate is the mass flow rate associated with flue gas with a downward velocity component in this upward firing configuration. The reverse flow ratio at each elevation is the ratio of the total reverse flow rate to the total mass flow rate exiting the furnace. Even though the flue gas flow rate in the furnace decreases as the natural gas fuel is replaced by the H_2 fuel, the reverse flow rate increases as the H_2 mole percentage in the fuel increases up to ~75 percent, then decreases as the H_2 mole percentage in the fuel increases from 75 to 100 percent. However, the reverse flow ratio increases continually as the H_2 mole percentage in the fuel increases from 0 to 90 percent, then maintaining the similar rate when the H_2 mole percentage from 90 to 100 percent. Increased reverse flow ratio contributes to a reduction in flame temperature, thereby limiting the NOx emissions.

Figure 11 shows the average reverse flow temperature and average reverse flow O_2 concentration vs. furnace height as a function of H_2 mole percentage in the fuel. The increased average reverse flow temperature with increasing H_2 mole percentage is much smaller than the increase in the adiabatic temperature. Also, the O_2 concentration of the recirculated gases reduces with increasing H_2 content. The combination of increasing the reverse flow ratio, reduced O_2 in the recirculating gases, and limited temperature increase of the recirculating gases with increasing H_2 content in the fuel, contributes to the limited NOx emission increase, followed by the NOx emission decrease shown in Figure 9.

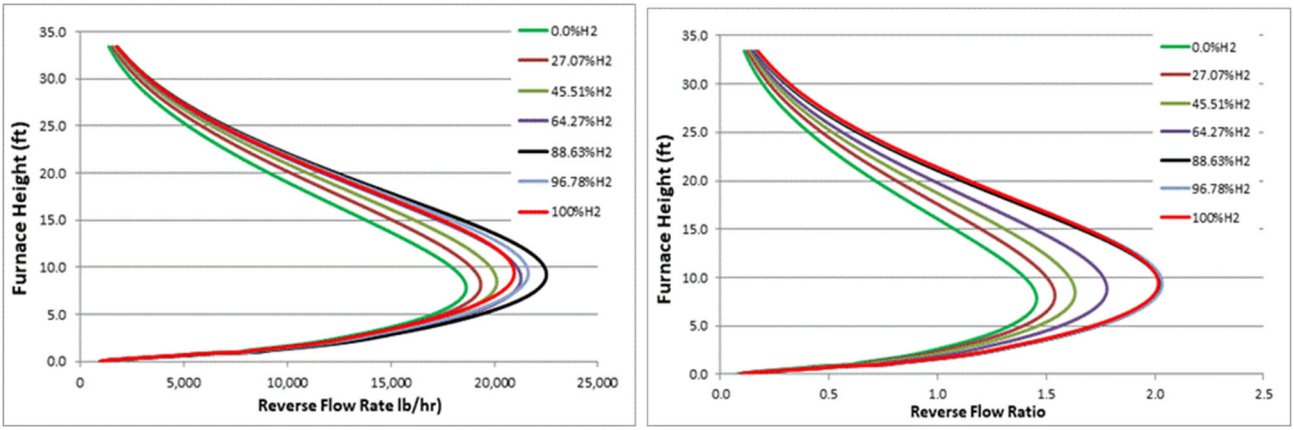

Figure 10: Reverse flow rate and reverse flow ratio vs. furnace height as a function of H_2 mole percentage in the fuel

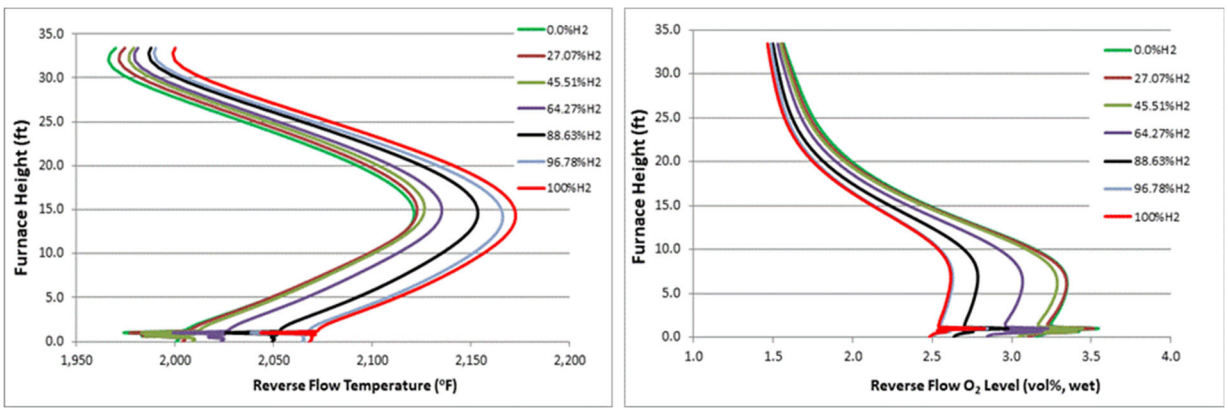

Figure 11: Average reverse flow temperature and average reverse flow O_2 concentration verse furnace height as a function of H_2 mole percentage in the fuel

3.3 Results of Power Boiler Simulations

The overall simulation results of the radiant section of the power boiler are listed in Table 1. The predicted model exit temperature is similar for natural gas fuel (1882°F) and for H_2 fuel (1881°F) even though ~ six percent more heat loss is predicted for H_2 fuel as shown in Table 1. NO_x emission is expressed in different forms. The biggest increase (53.5 percent) is in the form of NO_x ppmv @ 3 percent O_2 dry; the smallest increase (5.3 percent) is in the form of lb-NO_2/MBtu based on fuel HHV. Since the firing rate based on LHV is kept the same as natural gas fuel is replaced by H_2 fuel, NOx emission increase (12.3 percent), expressed as lbs-NO_2/MBtu, based on fuel LHV, is more reasonable to assess NO_x increase when the hydrocarbon fuel is replaced by the H_2 fuel.

Figure 12 shows the iso-thermal surfaces (3200°F and 2900°F) for natural gas fuel firing and H_2 fuel firing. These iso-surfaces are colored by the O_2 concentration. Elevated temperatures for the H_2 fuel are shown when comparing iso-thermal surface of 3200 °F and 2900°F for natural gas fuel and for H_2 fuel. The elevated gas temperature is clearer when plotting the mass weighted average temperature as a function of furnace height as shown in Figure 13. In the burner region, a temperature increase of about 100°F, much lower than 350 °F adiabatic temperature increase, is observed for H_2 fuel combustion in comparison with natural gas fuel combustion. This mainly is due to more radiation heat loss for the H_2 fuel combustion.

Table 1: Overall Predicted Results

Model Exit Plane	100% Natural Gas	100% H_2 Fuel	
Average Flue Gas Temperature (°F)	1882	1881	
Average O_2 (vol%, wet)	2.30	2.13	
Average O_2 (vol%, dry)	2.82	3.15	
Average CO (ppmv, wet)	2.4	< 0.1	
Average CO (ppmv, dry)	2.9	< 0.1	NOx Increase
Average NO_x (ppmv, wet)	155.5	194.2	24.9%
Average NO_x (ppmv, dry)	190.6	287.3	50.7%
Average NO_x (ppmv) @ 3% O_2 dry	188.8	289.8	53.5%
NO_x Emission (lb-NO_2/hr)	420	471	12.1%
NO_x Emission (lbs-NO_2/MBtu Based on HHV)	0.227	0.239	5.3%
NO_x Emission (lbs-NO_2/MBtu Based on LHV)	0.252	0.283	12.3%

Predicted Heat Duty	100% Natural Gas	100% H_2 Fuel
Heat Absorbed by Water Wall (MBtu/hr)	816.6	876.7
Heat Absorbed by Roof (MBtu/hr)	9.8	9.9
Heat Absorbed by Platen SH (MBtu/hr)	74.6	78.9
Heat Absorbed by Finishing SH (MBtu/hr)	97.7	94.4

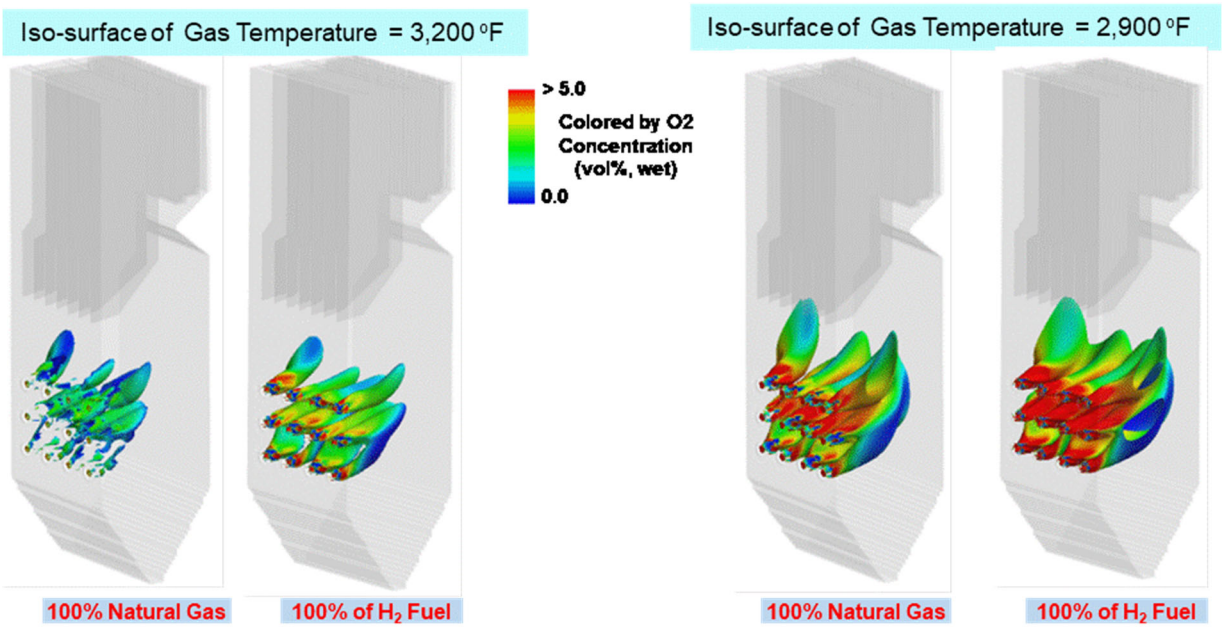

Figure 12: Iso-thermal surface of 3200°F and 2900°F when burning natural gas and H_2 fuel in a power boiler

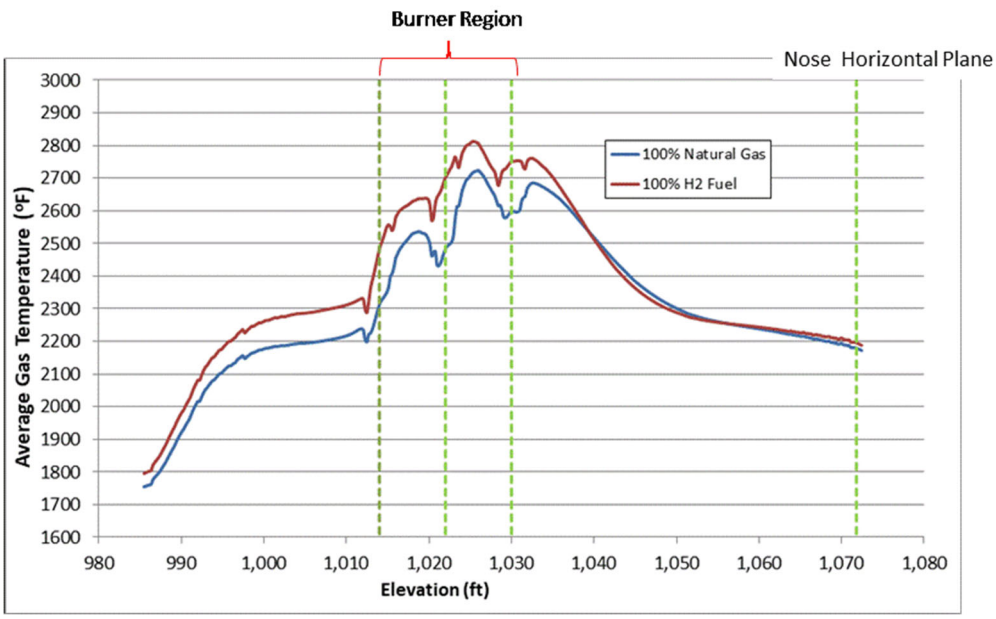

Figure 13: Mass weighted average gas temperature as a function of furnace height

Due to unstaged firing and limited flue gas recirculation in this power boiler, this increased gas temperature for H_2 fuel combustion results in increased NO_x concentration for H_2 fuel as shown in Figure 14. NO_x concentration is also plotted as NO_x flow rate as a function of furnace height in Figure 15. The majority of NO_x is formed in the burner region as shown in Figure 15. Figure 16 shows the net heat fluxes predicted for natural gas fuel combustion and H_2 fuel combustion. Increased radiation heat loss for H_2 fuel combustion results in elevated net heat flux for the waterwall in the burner region, as shown in Figure 16.

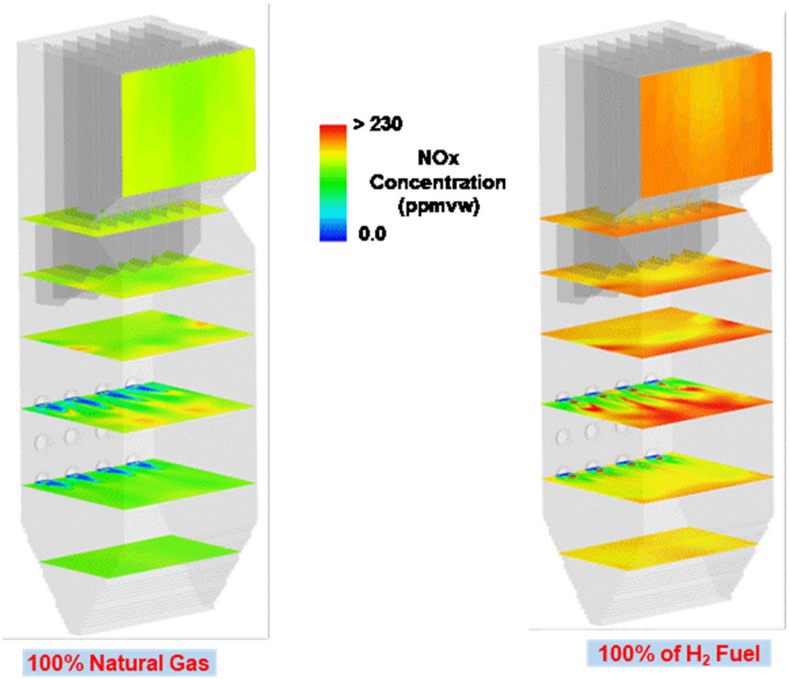

Figure 14: Predicted NO_x concentration when burning natural gas and H_2 fuel in a power boiler

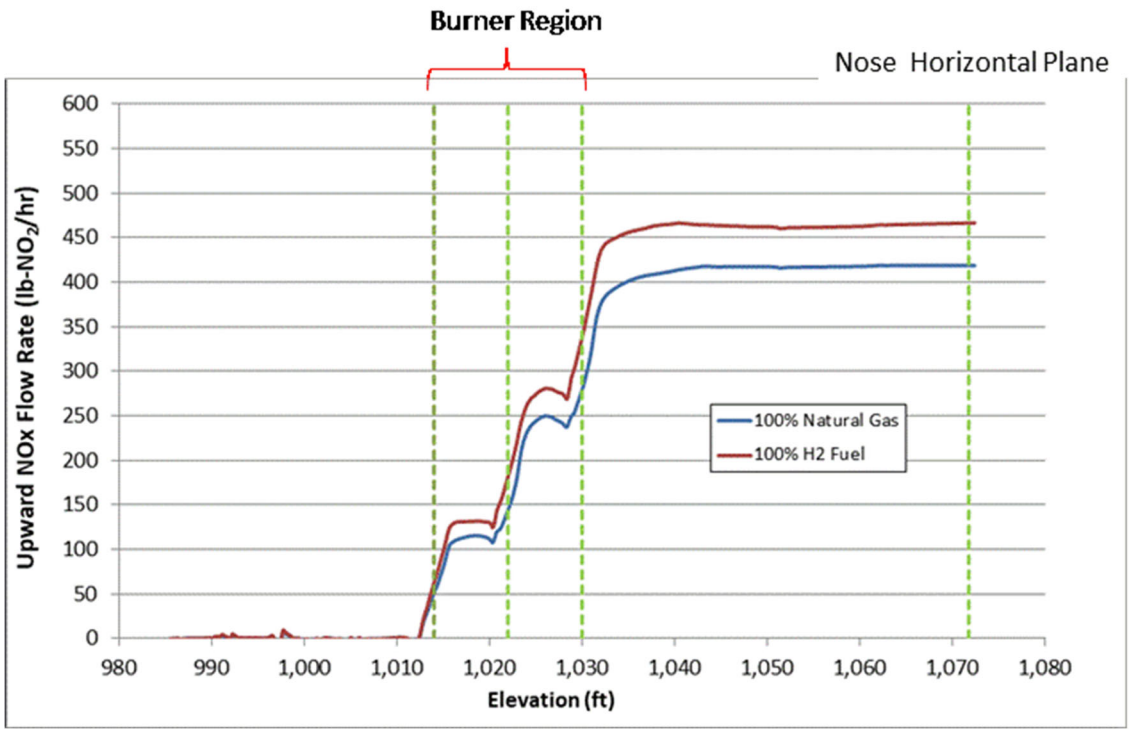

Figure 15: NO_x flow rate as a function of furnace height

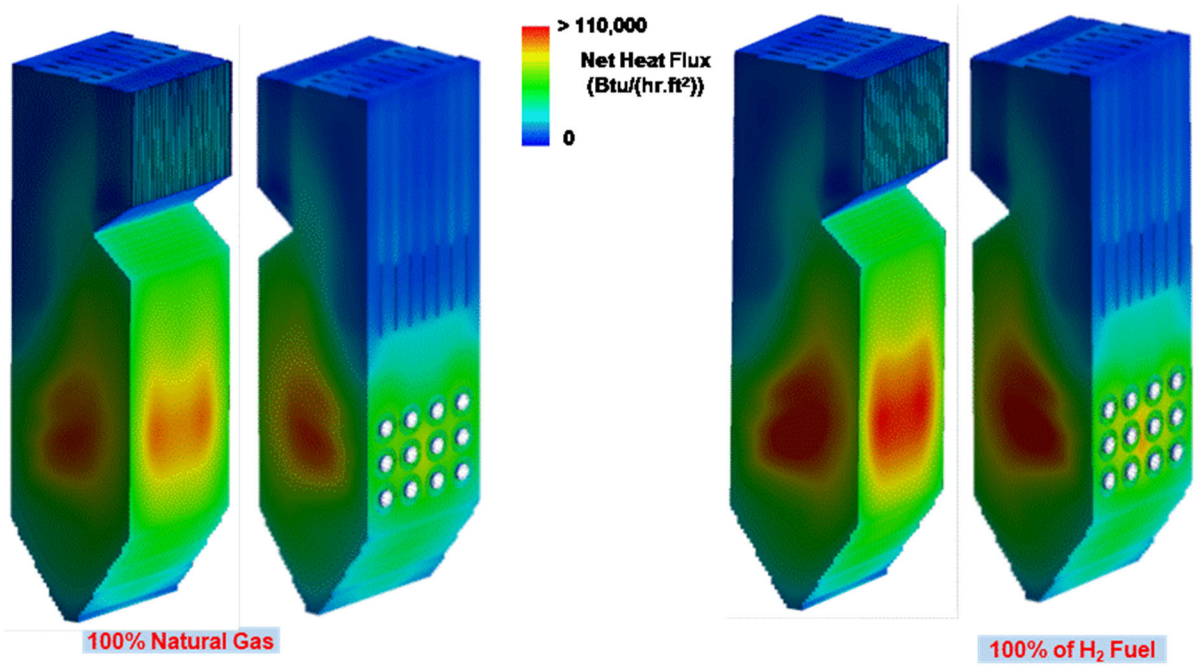

Figure 16: Predicted net heat flux for burning natural gas and for burning H_2 fuel

4 Conclusions

The results of the model-based evaluation described in this paper show that the NOx emission impacts of increasing the H_2 content in fuel blends with natural gas will generally depend on the firing configuration. The assumption that NOx emissions will always be higher with increasing H_2 content may be in error. The results shown here indicate that for an ethylene furnace, where there is significant bulk recirculation of cooled flue gases, the NOx emissions may be significantly non-linear as a function of H_2 content, even decreasing at high H_2 content. This behavior is likely associated with heavily fuel staged burners, where increasing H_2 content in the fuel increases the burner momentum, which has a positive impact on recirculation of cooled flue gases into the base of the flame.

On the other hand, the increasing flame temperature, and shorter flames produced with higher H_2 content fuel gas will typically yield increased heat transfer in the radiant section of a boiler or furnace, in the absence of the use of forced flue gas recirculation or significant combustion modifications accompanying the fuel change. A three-dimensional CFD model, which can accurately represent radiation heat transfer, including an adequate combustion model with good finite rate combustion and NOx kinetics, can be a very useful tool for evaluating the impacts of increasing the H_2 content of fuel blends, on both NOx emissions and thermal performance in industrial furnaces and boilers.

References

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