

# 3.2

## Combustion Strategies for Syngas and High-Hydrogen Fuel



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### 3.2-1 Introduction

The technical challenges surrounding syngas and hydrogen fuel combustion have been outlined in section 3.1. Given the issues presented there, various options can be considered for combustor design and operation. First, it is critical to define the type of combustion system that will be used. There are two broad categories: diffusion flame combustors, and premixed combustors. These are described below, but before discussing the combustion strategies, it is useful to review how NO<sub>x</sub> pollutants are formed.

### 3.2-2 NO<sub>x</sub> Formation

There are several routes to form NO<sub>x</sub> pollutants and these may be broadly catalogued as thermally-generated, flame-generated, or fuel-bound NO<sub>x</sub>. Different authors use different names to catalogue these mechanisms and there is still continuing research to understand the most prominent mechanisms at ultra-low NO<sub>x</sub> conditions. For example, in hydrogen fueled systems, the prominence of H radicals may contribute to NO<sub>x</sub> in a manner that is different than in systems fueled by natural gas.<sup>1</sup>

Thermal NO<sub>x</sub> is formed by oxidation of nitrogen in air and requires sufficient temperature and time to produce NO<sub>x</sub>. A rule of thumb is that below approximately 1700K, the residence time in typical gas turbine combustors is not long enough to produce significant thermal NO<sub>x</sub>. Where temperatures higher than 1700K cannot be avoided, it is necessary to limit residence time to control NO<sub>x</sub> formation, which favors very short combustor designs. Thermal NO<sub>x</sub> production also increases with the square root of operating pressure, making it more difficult to reduce in higher-pressure aeroderivative gas turbines.

As the name implies, flame-generated NO<sub>x</sub> occurs in the flame front, created on the short time scale associated with primary combustion reactions. There are a variety of chemical mechanisms involved, all linked to intermediate combustion species that exist only in the reaction zone of the flame. It is important to understand that in practical combustors, the reaction zone is just a small portion of the total combustor volume –most of the combustor volume is sized to complete the relatively slow approach to equilibrium products (notably CO to CO<sub>2</sub> oxidation). Thus, residence time in the whole combustor does not affect the flame-generated NO<sub>x</sub> produced – a significantly different behavior compared to thermal NO<sub>x</sub>. A convincing demonstration of this point was presented by Leonard and Stegmaier<sup>2</sup> who studied multiple flame holders, operating conditions, and residence times from 2 to 100 milliseconds, demonstrating that the flame temperature alone (not residence time) determined the NO<sub>x</sub> production for emissions under 10 ppmv. Fig. 1, is useful to estimate the flame NO<sub>x</sub> produced at a given flame temperature, accounting for ideal, and “poor” premixing (not carefully defined in note 2). Note that the effect of poor premixing raises the NO<sub>x</sub> levels by as much as a factor of three. These data were recorded in turbulent flames, where combustion products are mixed with the fresh reactants right at the flame. It has been suggested that other combustion configurations, without significant stirring between the flame front and products, may reduce the flame generated NO<sub>x</sub>.<sup>3</sup> This may be the basis for NO<sub>x</sub> reductions reported in the Low-Swirl Combustion section.

Finally, fuel-bound NO<sub>x</sub> is produced by nitrogen species in the fuel reacting with air during combustion. For coal syngas, the most prominent fuel nitrogen species is ammonia, generated during gasification from nitrogen compounds in coal. The ammonia should ideally be removed from the fuel before entering the combustor, or it will be converted to NO<sub>x</sub> by most combustion strategies. Where this is not possible, rich-lean strategies have the most potential to reduce NO<sub>x</sub> pollutants. In this approach, combustion is first carried out under fuel-rich conditions, followed by completing combustion under fuel lean conditions. In fuel rich conditions, with sufficient residence times, the ammonia can be reduced to nitrogen and water, rather than

atmospheric oxygen. A number of studies have been conducted to evaluate rich-lean combustion as an approach to reducing fuel bound NOx. These studies have shown as much as 95% of the fuel ammonia can be reduced to nitrogen and water using rich-lean combustion, with the remaining 5% converting to NOx.<sup>4</sup> Untreated syngas ammonia concentrations can exceed 1000ppm, where even 5% conversion would lead to 50ppm NOx, which is well above desired emissions levels. Thus, it is desirable to remove fuel ammonia during gas cleanup, rather than rely on combustion techniques to reduce it to water and nitrogen.

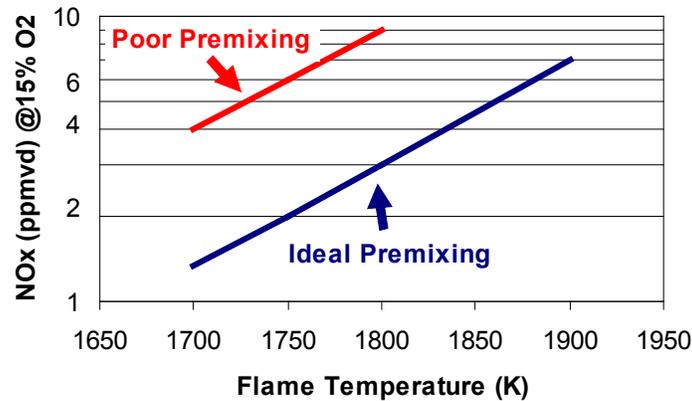


Fig. 1. NOx emissions, adopted from Leonard and Stegmaier.

Source: See note 2.

### 3.2-3 Diffusion Flame Combustion

In this style of combustion, fuel and air are introduced in separate passages, and the flame is stabilized where the fuel and air streams mix. Combustion reactions are typically so fast that fuel and oxidant consumption is limited by transport to the reaction zone (i.e., diffusion), and the reaction proceeds locally at nearly stoichiometric conditions. The Lewis number (Le) describes the ratio of thermal transport to species transport from this reaction zone. Where  $Le = 1$ , the temperature in the reaction zone will equal the adiabatic flame temperature because thermal energy diffuses away as fast as the reactants are supplied. The fuel species in hydrocarbon combustion typically have fuel Lewis numbers ( $\alpha_{mix} / D_{j_{fuel}}$ ) in the range of 0.9 to 1.2, meaning that diffusion flame combustors will have flame temperatures near the adiabatic flame temperature. These temperatures are high enough to oxidize nitrogen in air, producing appreciable NOx pollutants. Hydrogen itself has a fuel Lewis number as low as 0.4, making it even more difficult to reduce NOx because the peak laminar flame temperatures are higher than adiabatic due to differential diffusion effects. The effect of fuel Lewis number on flame temperature has been observed experimentally as well as with direct numerical simulations (DNS).<sup>5</sup>

Because of their high flame temperatures, diffusion flame combustors require some method to achieve low-NOx performance. An obvious technique is to dilute the fuel, lowering the adiabatic flame temperature. A common diluent is steam, which can both lower the flame temperature, and reduce the production of non-thermal NOx. The hydroxyl radical OH is increased by the presence of additional water, and these radicals favorably scavenge HCN fragments which might otherwise produce NOx. Steam dilution is already used on IGCC applications, but it is not completely desirable. The extra energy that is needed to make steam from water is not recovered in the turbine expansion, penalizing cycle efficiency, (but raising power output from the added mass flow). The additional steam in the exhaust produces a modest increase in the turbine nozzle heat transfer, raising metal temperatures. The protective thermal oxide layers in turbine material sets can be affected by increased moisture levels. Finally, steam consumption by stationary turbines should be minimized to conserve water resources. For these reasons, any further development of diffusion flame combustors for IGCC applications would ideally use nitrogen from the air separation plant, rather than steam. The amount of nitrogen available for flame dilution is established by the engine cycle and the ASU, and it can be shown that for example, hydrogen could be diluted up to about 50% with nitrogen in a typical IGCC configuration. Unfortunately, this level of dilution produces an adiabatic flame temperature around 2025 K, which is still too high for ultra-low NOx performance.

Given the dilution limit on adiabatic flame temperature, it is important to consider other methods to reduce the diffusion flame temperature. As noted above, the diffusion flame temperature is set by the ratio of thermal diffusion away from the reaction zone to heat generated by reactants. If the reaction zone is “strained” by fluid shear, it is possible to change the balance between diffusion and reaction in the reaction zone, changing the flame temperature. Strongly sheared flows can locally extinguish the flame, providing opportunity for fuel air mixing before combustion is initiated elsewhere. This raises the possibility that strong shearing could be used to make a diffusion flame combustor behave more like a premixed combustor. The required levels of shearing (known as “stretch” or “strain”) have not been fully characterized. These concepts are discussed in the Highly-Strained Diffusion Flame Combustion section.

### 3.2-4 Lean Direct Injection

Lean Direct Injection (LDI) combustion was developed as a low NO<sub>x</sub> alternative to Lean Prevaporized Premixed (LPP) combustion for aircraft gas turbines, where the inherent flashback and dynamic instability concerns of LPP combustion are considered too great of a risk for flight application. In LDI combustors, liquid fuel is directly injected into the combustion chamber, where it is mixed with air in the shortest possible distance. The intent is to provide an essentially lean premixed fuel/air mixture that burns in a low-NO<sub>x</sub> flame, similar to LPP combustors, which are discussed in the Premixed Combustion section below.<sup>6</sup>

Low-NO<sub>x</sub> performance is compromised in an LDI combustor if the fuel and air are not perfectly mixed before combustion occurs, creating regions with higher fuel content that burn hotter and generate more NO<sub>x</sub>. Similarly, the mixture may burn upstream of the premixed zone in a diffusion flame, with combustion occurring at stoichiometric conditions that result in higher temperatures and NO<sub>x</sub> production. Nevertheless, flashback and auto-ignition concerns are nearly eliminated in LDI combustors, and they can operate over a wide turndown range with a high degree of static and dynamic stability using a wide range of fuels.

The desire to burn high-hydrogen fuels in gas turbines used for power applications raises similar concerns of flashback and instability when operating in the Lean Premixed mode of combustion, so LDI combustors seem to be a natural fit for burning these fuels in a low NO<sub>x</sub> gas turbine system. To demonstrate the potential of LDI combustors, researchers at NASA Glenn have recently studied various low NO<sub>x</sub> LDI concepts for pure hydrogen combustion in aircraft gas turbine combustors.<sup>7</sup> Five separate injector concepts from different manufacturers were tested at aircraft gas turbine conditions (4.8 – 13.6 atm,  $T_{in} = 600 - 1000$  °F). At low combustor exit temperatures, it was possible to achieve very small NO<sub>x</sub> levels (~1 ppmv, wet, uncorrected). NO<sub>x</sub> emissions were primarily controlled by lowering equivalence ratios to limit combustion temperatures, and no hydrogen dilution cases were considered.

One of the tested injectors at NASA Glenn was similar to those used in current IGCC gas turbines that burn syngas, where fuel is injected axially into a swirling airflow. Although this injector was very robust, it produced substantially higher NO<sub>x</sub> than the other tested injectors. Some of the other tested injectors were similar to those studied recently at GE Energy, where multiple fuel jets were injected at an angle into a central air jet.<sup>8</sup> Their results show that more fuel injection ports per air jet reduce NO<sub>x</sub> emissions due to higher fuel jet momentum and mixing. Increasing the number and decreasing the size of the air jets is shown to reduce NO<sub>x</sub> by reducing the length of the combustion zone, although this comes at the expense of increased combustor pressure drop. Similar injector configurations studied at NASA Glenn had better NO<sub>x</sub> emissions, due in part to the shortened combustion zone. However, in some cases, this also led to overheating problems and injector failure, since the combustion zone was located much closer to the fuel and air injectors.

Pressure drops in the NASA Glenn injectors were sometimes very large (4-25%). Redesign and optimization for power gas turbines could reduce these pressure drops. In addition, large pressure drops may have been required to reduce the flashback or flameholding potential in those injector designs that operated more in a premixed combustion mode than a diffusion combustion mode. As the injectors were tested on pure hydrogen, dilution with nitrogen will reduce flame speeds and may decrease the necessity for large injector pressure drops and high air velocities to avoid these issues.

### 3.2-5 Highly-Strained Diffusion Flame Combustion

Though not discussed explicitly in the above studies, successful LDI diffusion flame combustors use jets of fuel and air that introduce high strain rates in the combustion zone. In a pure diffusion flame, strain rate can be quantified by measuring or calculating the velocity gradients in the mixing flow field. In regions of high strain and fluid shear, mixing rates and bulk transport rates are faster than chemical reaction rates, thus local reactions are not allowed to go to completion before the flow carries the combustion radicals away from the reaction zone. The net result of this process is a reduction in peak flame temperature of a highly strained flame, which in turn reduces thermal NO<sub>x</sub> production. It should be pointed out, however, that thermal NO<sub>x</sub> is not only a function of temperature, but also of flame residence time and O-atom concentration in the reaction zone. Increasing the flame strain also tends to reduce the residence time in the flame, but it also can increase the O-atom concentration in the flame by an order of magnitude. This effect is shown in Figure 2, where intermediate strain rates tend to increase the production rate of NO due to the increased O-atom concentration, while at high strain rates, the reduction in flame temperature overcomes the influence of the O-atom concentration, and NO production rates are reduced. This points to increased strain rates as a possible path to reducing or effectively eliminating thermal NO<sub>x</sub> in a diluted diffusion flame, where dilution of the fuel alone does not reduce flame temperatures enough to satisfy ultra-low NO<sub>x</sub> emission goals.

Increased strain rates are typically attained by increasing the fuel and/or air jet velocities to increase fluid shear, though at the expense of increased combustor pressure drop. In addition, the static stability of the flame is a strong function of these jet velocities, where too high of a jet velocity could cause the flame to blowout. Thus, flame stability concerns place limits on allowable levels of flame strain, particularly for diluted high-hydrogen content fuels, since flameholding ability is closely linked to the flame speed of the fuel/air mixture, which decreases as more diluent is added to the fuel stream. From this perspective, impinging fuel and air jet injector configurations<sup>9</sup> hold an advantage over co-axial jet configurations, as forced mixing of the fuel and air should improve the flameholding abilities of these diffusion flames.

Much more study could be done in this area to determine injector configurations that maximize flame strain while minimizing stability and combustor pressure drop concerns. In addition, the effect of strain rate on NO<sub>x</sub> emission from diffusion flames has only been partially quantified for simple diffusion flames, and there are no such studies in practical LDI-type diffusion flame combustors using hydrogen, syngas, and/or fuel diluents. Other areas requiring further study include the effects of increased flame strain on combustion efficiency and on in-flame NO<sub>x</sub> production mechanisms.

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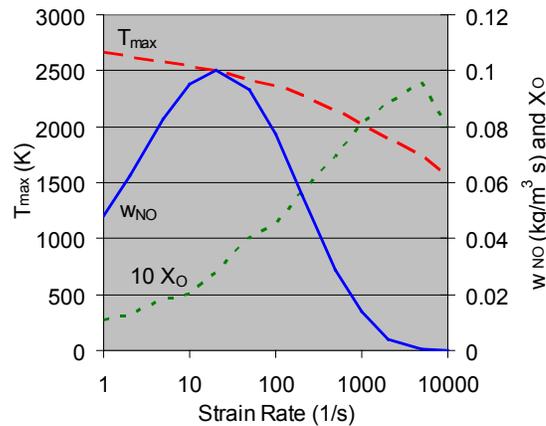


Fig. 2. Strain rate effects, adapted from Sanders et. al.  $w_{\text{NO}}$  = NO formation rate,  $X_{\text{O}}$  = O-atom mole fraction,  $T_{\text{max}}$  = peak temperature

Source: Sanders, J. P. H., Chen, J.-Y., and Gokalp, I., "Flamelet-Based Modeling of NO Formation in Turbulent Hydrogen Jet Diffusion Flames," *Combustion And Flame*, Vol. 111, pp. 1-15, 1997.

### 3.2-6 Premixed Combustion

As the name implies, premixed combustion is accomplished by mixing the fuel and air upstream of the flame. The fuel-air ratio normalized by the stoichiometric value is known as the equivalence ratio  $\phi$ , and in many practical premixed turbine combustors, has a value of slightly more than 0.5. Thus, there is approximately  $\frac{1}{2}$  the fuel needed to burn all the air, or conversely twice as much air as needed to burn all the fuel. The excess air serves to dilute the combustion and keep the flame temperatures low enough to avoid thermal NO<sub>x</sub> formation. While the concept of premixed combustion is simple and effective at reducing NO<sub>x</sub>, it also has drawbacks. The combustor must operate in a very narrow range of equivalence ratio to avoid blowout at (typically)  $\phi < 0.5$ , and increasing NO<sub>x</sub> formation for  $\phi$  somewhat greater than 0.6. The combustor controls must include some form of staging, since the range of desired exit temperatures usually cannot be achieved with such a small range of  $\phi$ . For example, if four fuel injectors are used in a combustor, it is possible to reduce the heat input 50% keeping two injectors operating, but turning two off. The difficulty with this approach is that the air flow from inactive injectors can quench the boundary of the flame from operating injectors, raising CO emissions, but this can be addressed with good aerodynamic design. Staging in this manner is used on commercial engines.<sup>10</sup> Beyond simply de-activating injectors, staging is also accomplished by operating some injectors at slightly richer equivalence ratios, to improve flame stability. This can also be accomplished using "pilots" on individual injectors. The pilot flame is typically supplied with some air for partial premixing, and the pilot fuel circuit is controlled to achieve stable combustion at the lowest possible NO<sub>x</sub> emissions, as described in the following section.

### 3.2-7 Tuning and Combustor Control

Balancing the fuel delivery among various fuel circuits to meet operating requirements is known as "tuning" and has become a critical part of both commissioning and operating low-emission gas turbines. Various strategies have been used, or are being developed so that the emissions targets can be met with stable combustion. Because combustion stability is affected by inlet temperature and fuel composition, tuning may need to be adjusted to accommodate ambient environment temperatures and even fuel composition. In addition to controlling the fuel split, for some turbines, tuning may include adjustment of compressor inlet guide vanes or bleeding compressor flow<sup>11</sup>. This allows an adjustment of the combustor air flow at fixed compressor speed, providing another tuning option even on single-speed (synchronous) gas turbines.

It is important to understand that turbines must be able to contend with requirements for load rejection while low-emission combustors operate near the blowout condition. Without careful development, cutting the fuel during load rejection can lead to flame blowout, requiring (sometimes) unacceptable time to re-light and establish power, or making the engine unable to meet grid requirements. An interesting account of the development of combustor and control system required to meet stringent rejection requirements is given in the references.<sup>12</sup>

On some engines, fast acting valves are used to enhance lean-blowout performance<sup>13</sup> and allow operation right near the limits of stable combustion. A more advanced concept is to modulate the fuel to counteract combustion oscillations, usually called active combustion control. Active control has been studied in many research projects<sup>14</sup>, but has only been deployed on one test engine<sup>15</sup> and on one commercial engine installation<sup>16</sup> to date.

An important aspect of combustion tuning and control is diagnosing conditions in the combustor so that the control system can respond to maintain stable, low-emission operation. For example, it is possible to improve engine operation by monitoring combustion performance from available engine sensors.<sup>17</sup> A number of recent papers have shown the potential of using flame optical signals, acoustic signals, or flame ionization to monitor and control the combustion process.<sup>18</sup>

### 3.2-8 Oxy-Fuel Combustion

As noted in section 1.3.1, advanced engine cycles using oxy-fuel combustion have been proposed as a means of capturing CO<sub>2</sub> from engine operation. These oxy-fuel cycles require a different approach to combustor design because the combustion is ideally operated at stoichiometric conditions – having just enough oxygen to completely oxidize the fuel. Oxygen is produced from air separation, such that any excess oxygen is produced with an accompanying penalty to the overall cycle efficiency. In addition, after the water is condensed from the exhaust, any excess oxygen should be eliminated from the compressed CO<sub>2</sub> to avoid corrosion in handling the CO<sub>2</sub> gas. For these reasons, the combustor design must achieve very high combustion efficiency at conditions with little excess oxygen. This requirement places a premium on achieving high levels of mixing uniformity in the combustor, because even modestly unmixed fuel stream will be starved for oxygen. It should be noted that boiler designs also ideally operate near stoichiometric, but typically use 1-3% excess oxygen, and have relatively long residence times to complete fuel oxidation. For the oxy-fuel turbine, the excess oxygen would ideally be lower, with much shorter residence times (~30ms) to avoid excessively large pressurized combustion chambers. Oxy-fuel combustion for power cycles has been studied in a number of papers.<sup>19</sup> The easiest combustion strategy is to employ a diffusion flame combustor. The stability and simple operation of diffusion flame systems make them appealing for oxy-fuel systems. There is no need to control NO<sub>x</sub>, since the products are sequestered, and there is otherwise little nitrogen in the combustor. Even without sequestration, the peak flame temperature in diffusion flames can be controlled by the level of diluent added, thereby avoiding NO<sub>x</sub> formation. Nevertheless, a potential advantage of premixed combustion is that premixing the fuel and oxidant can reduce the unmixed streams of fuel and oxygen that are created in diffusion flame systems where relatively small fuel jets must penetrate and mix in the large combustion volume. There is relatively little fundamental data on premixed oxy-fuel flames diluted by water or CO<sub>2</sub><sup>20</sup> such that proposed designs must include some margin with respect to fundamental issues like flame speed.

### 3.2-9 Notes

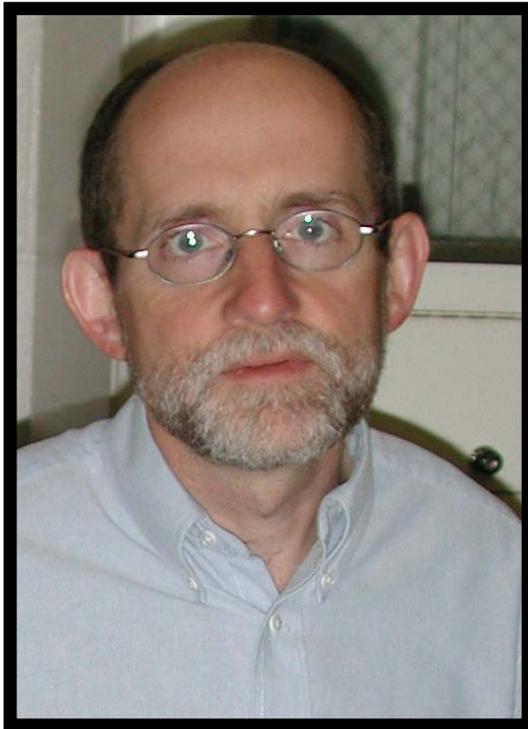
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Geo Richards received his Ph.D. in mechanical engineering from Purdue University on the subject of gas turbine combustion. Since coming to the National Energy Technology Laboratory in 1988, he has conducted research on various topics in thermal science and energy production, with a particular emphasis on combustion dynamics. He currently leads the Energy System Dynamics Focus Area, providing technical direction for research groups investigating turbine combustion, carbon dioxide capture, high-temperature fuel cells, fuel processing, and stationary reciprocating engines. In addition to conducting his own research, Dr. Richards' responsibilities include developing and executing cooperative research agreements with private industry and academia, and evaluating proposed concepts related to energy conversion. He also serves as a research advisor for both graduate and post-graduate investigators visiting NETL from academic institutions.



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