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# Fundamentals and Simulation of MILD Combustion

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## 1. Introduction

### 1.1 Introduction and definitions

There is a continual demand to develop combustion systems that lead to reductions in pollutant emissions and increases in energy efficiency, thereby reducing fuel consumption. It is well known that thermal efficiency may be increased by preheating of the reactants (Özdemir & Peters, 2001). An immediate drawback of preheating techniques is that the increased peak flame temperature results in increased NO<sub>x</sub> emissions. Alternatively, to reduce NO<sub>x</sub> the aim is to reduce peak temperatures, which is often accompanied by an increase in other emissions, for instance carbon monoxide (CO) (Hamdi et al., 2004). It is this conflict in requirements which makes the combination of low emissions, high thermal efficiency burners difficult to design.

A significant development towards combustion which offers both low emissions and high efficiency was found when exhaust gases are recirculated into the reaction zone (Choi & Katsuki, 2001; Hasegawa et al., 1997; Katsuki & Hasegawa, 1998). The heat from the exhaust gases is recovered, increasing system efficiency and also preheating of the reactants. A low oxygen content hot artificial air mixture is obtained where in the exhaust gases act as an inert diluent which basically serves to increase the thermal mass of the system and to reduce fuel consumption. It is this combination of using high temperature and high dilution which is the basis of the Moderate or Intense Low oxygen Dilution (MILD) combustion regime.

It has been suggested that there is an excellent potential for the use of recirculation of exhaust gases to exploit the simultaneous heating and dilution effects to achieve improvements in both the emissions and efficiency, narrowing the gap between the two objectives of low pollutant emissions and fuel savings (Katsuki & Hasegawa, 1998).

In this field, a bewildering number of different terms are used to describe processes which are very similar in nature. While the descriptive titles attempt to highlight some of the subtleties of the various systems, overall many share the same or at least very similar, underlying principles. The sheer number of terminologies which are in use leads to many being misinterpreted or being confused for another. Rather than getting entangled in the twisted web of seemingly endless acronyms and overly descriptive names, for the sake of simplicity, in this discussion the processes will be segregated into one of two groups; preheated air combustion or MILD combustion – which itself being a subgroup of the preheated group.

To some, preheated air combustion refers to a highly specific combustion regime with completely unique and characterisable features. In essence however, the origins and fundamental principle of preheating is the heating of the reactants prior to combustion; which is done by means of recirculation, recuperation or external heating. While there are very important physical differences in the implementation, and in fact in the chemical processes too, preheating will be used to describe any system where the reactants are preheated. Although this may seem an oversimplification, it avoids getting bogged down in the details where this is not necessary for the issue at hand.

Preheating combustion air by means of recovering heat from the exhaust gases is recognised as one of the most effective ways of increasing the thermal efficiency of a combustion system (Borman & Ragland, 1996; Weber et al., 2000; Wüning, J.A. & Wüning, J.J., 1997). The use of such exhaust gas recirculation (EGR) has been developed since the 1970's as a way of increasing furnace efficiency, and subsequently minimising fuel consumption (Weber et al., 2000). The main drawback of increasing thermal efficiency with EGR is there can be an increase in temperatures, leading to increases in NO<sub>x</sub> emissions. Depending on the implementation, recirculation of exhaust gases increases the content of inert in the mixture (Wüning, J.A., & Wüning, J.J., 1997).

In the extreme case of significant recirculation of combustion products, where the amount of inert introduced by the recirculation is sufficient to alter the structure of the reaction zone, is where the MILD combustion regime lies. It can therefore be said that there is a close resemblance between preheating processes and MILD combustion. As such, it may be concluded that MILD combustion is a subset of the more general preheating principles, the difference arises from the dilution effects associated with the MILD regime. A further difference between the two is the extent of the preheating. To achieve MILD combustion conditions the temperature prior to combustion must exceed the auto-ignition temperature of the mixture, and is therefore classified as being highly preheated (Choi & Katsuki, 2001; Katsuki & Hasegawa, 1998).

The exact definition of the amount of preheating and recirculation required to move from a simple preheated mode to MILD combustion is somewhat ambiguous, and this is where several different combustion regimes have been identified. One attempt at making a definition of MILD combustion has been made by Cavaliere & De Joannon (2004): "*A combustion process is named MILD when the inlet temperature of the reactant mixture is higher than mixture self-ignition temperature whereas maximum allowable temperature increase with respect to inlet temperature during combustion is lower than mixture self-ignition temperature (in Kelvin)*". Cavaliere and De Joannon (2004) claim that this definition of MILD combustion is unambiguous because the conditions are well defined and univocal – however, it is based on the theory of a well-stirred reactor (WSR) and is therefore not particularly well generalised. An alternative criterion for MILD combustion has been suggested based on the temperature variations in the flow field by Kumar et al. (2002), but no definitive were placed to indicate a general definition.

There are a number of combustion regimes that have been developed which exhibit very similar properties to MILD combustion, each having different names. While each implementation is slightly different, they all operate on very similar principles. Based on the observation that under certain conditions the MILD combustion process may result in no visible or audible flame it has been termed Flameless oxidation (FLOX) by Wüning, J.A. and Wüning, J.J., (1997). On a related theme, some similar names are flameless combustion (Cavaliere & De Joannon, 2004), colourless combustion (Weber et al., 2000) or invisible

flames (Choi & Katsuki, 2001). Alternatively, other names have been derived using the requirement of preheating; high temperature air combustion (HiTAC); high temperature combustion technology (HiCOT) (Cavaliere & De Joannon, 2004) or excess enthalpy combustion (Weber et al., 2000). There are subtle differences in each of these combustions technologies, but they all rely on the fundamental principle of the reaction taking place with high dilution levels and in a high temperature environment. For this reason, a single grouping of MILD combustion seems appropriate.

### 1.2 Principle of MILD combustion

The two basic conditions required for MILD combustion are high dilution and increased temperature of the reactants. Typically, both of these criteria are met by recirculation of the exhaust gases into the reaction zone. Entrainment of exhaust gases into the combustion zone is very important for the initiation of MILD combustion (Özdemir & Peters, 2001). Depending on the characteristics of the recirculation, it is possible to achieve various combinations of recirculation rates and temperatures, resulting in different combustion modes as shown in figure 1 (Wünning, J.A. & Wünning, J.J., 1997).

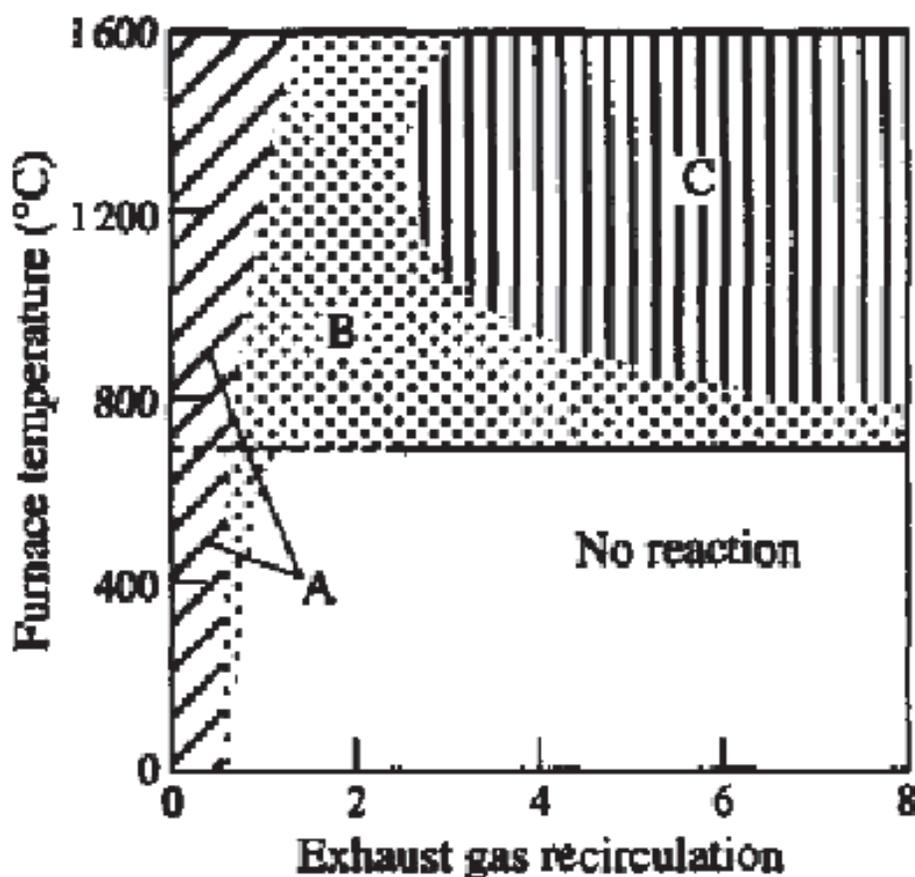


Fig. 1. Stability limits of conventional and MILD combustion (Wünning, J.A. & Wünning, J.J., 1997)

Along the horizontal (x-axis) of Figure 1 is exhaust gas recirculation, and is a measure of the proportion of exhaust gas to “clean” air. Mathematically, the recirculation rate may be written as:

$$K_v = \frac{\dot{M}_E}{\dot{M}_F + \dot{M}_A} \quad (1)$$

where  $\dot{M}$  refers to mass flow rate, and the subscripts represent; E: recirculated exhaust gas, F: fuel, A: "clean" combustion air. The vertical (y-axis) labelled "Furnace temperature" refers to the temperature of the reactants mixture.

Region A shows the domain of conventional flames, where the recirculation rate is less than ~30%. Such flames are stable. When the recirculation rate is increased, into region B, the resultant flame has been found to become unstable. If the temperature is below the self-ignition temperature (the horizontal line at ~700°C), these flames will extinguish. In region C, it has been found that if the recirculation rate is increased sufficiently, and the temperature is above that of self-ignition, that stable combustion results. It is this mode of operation that is known as the MILD combustion regime.

Figure 1 gives an indication of a fundamental difference between conventional and MILD combustion; the vast increase in the flammability limits (De Joannon et al., 1997), as well as improved flame stability limits (Hasegawa et al., 1997). The reaction at very low oxygen levels, and the improvement in flame stability under MILD conditions, is attributed to the temperature being above that of self-ignition, implying that the flame will always be sustained inside the furnace (Katsuki & Hasegawa, 1998).

By diluting the oxygen stream, the combustion reaction is more distributed, in turn distributing the heat release. With the heat release occurring over a larger volume temperature peaks are avoided, thereby thermal NO formation is largely suppressed (Wünning, J.A. & Wünning, J.J., 1997). Although the heat release occurs over a larger volume, the total is the same, implying that under MILD combustion conditions the heat release rate per unit volume is lower (Hasegawa et al., 1997).

### 1.3 Characteristics of MILD combustion

One of the most significant effects of dilution under MILD combustion conditions is that the extent of the reaction zone increases. This in turn has the effect of creating a far more uniform temperature distribution throughout the combustion region. In addition to the spatial uniformity of the temperature distribution, there is a significant reduction in the temporal fluctuations too – a reduction in the temporal temperature RMS of 98% has been reported (Hasegawa et al., 1997).

The distributed thermal field associated with MILD combustion also leads to the reduction of peak flame temperatures. By eliminating regions of high temperature the formation of NO<sub>x</sub> is largely suppressed. Ten-fold reductions in NO<sub>x</sub> emissions are possible with MILD combustion, and no specific region is identified where NO<sub>x</sub> is formed (Weber et al., 2000). Although the amount of NO<sub>x</sub> produced by MILD combustion is significantly less than conventional combustion systems, some NO<sub>x</sub> is still generated. Factors such as composition and mixing can affect the local combustion properties and thus the amount of NO<sub>x</sub> produced. For instance, the location of the inlet nozzle (hence the mixing process) drastically affects NO<sub>x</sub> emissions, and cannot be attributed to imperfect combustion since no unburned hydrocarbons are measured in the exhaust (Choi & Katsuki, 2001). Different diluents also influence the production of NO<sub>x</sub>, with much less NO<sub>x</sub> being formed with CO<sub>2</sub> dilution as compared to N<sub>2</sub>.

Despite the less intense reaction surrounding MILD combustion, both unburned hydrocarbons and carbon monoxide (CO) level are very low, or even not detectable, at the outlet of a MILD combustion furnace (Weber et al., 1999, 2000). The low emission of CO (< 100 ppm) suggests stable combustion conditions exist with such furnaces (Hasegawa et al., 1997). The temperature range encountered in MILD combustion also has significant advantages in the reduction of soot (Cavaliere & De Joannon, 2004).

To achieve stable combustion at the low oxygen levels associated with the very high recirculation rates of MILD combustion it is necessary for the initial temperature to exceed the auto-ignition temperature (Katsuki & Hasegawa, 1998). The method of achieving recirculation can be internal or external with regard to the combustor (Szegö et al., 2007). The recirculation, typically achieved with high velocity nozzles (Cavaliere & De Joannon, 2004), and the resultant reduction in oxygen concentration, lead to MILD combustion flames being associated with low Damköhler numbers (Katsuki & Hasegawa, 1998). In conventional flames, the chemical kinetics occurs much faster than the mixing, whereas under MILD conditions, the combustion is controlled by both the kinetics and the mixing (Milani & Saponaro, 2001). As an extension to this, in a furnace environment, MILD combustion may be likened to a well-stirred reactor (WSR) as the chemical time scales becoming larger in relation to the turbulence time scales (Plessing et al., 1998). The link between MILD combustion and a well Stirred Reactor may tend to indicate that this regime should be readily modelled, since a well stirred reactor is well defined. The interaction between the turbulence and the chemistry in describing the nature of MILD combustion is expected to play a significant role however (Katsuki & Hasegawa, 1998), and so the analogy to a WSR may not always be applicable.

One of the most noticeable characteristics of MILD combustion is the differences in visual appearance compared to other flames. When, operating in MILD conditions, furnaces have been described as “glowing”, with no actual flame visible (Weber et al., 2000). This type of description of the appearance of MILD combustion is very typical. MILD combustion is frequently described as appearing weak, with diffuse and distributed reactions zones (Katsuki & Hasegawa, 1998).

The visible emission from MILD combustion is measured to be two orders of magnitude lower than from conventional flames (De Joannon et al., 2000). In the case of methane, the predominate sources of colour in typical flames is due to C<sub>2</sub> and CH, both of which are formed at relatively high temperatures, which are avoided in MILD conditions, therefore the lack of colour (Cavaliere & De Joannon, 2004). Descriptions of combustors typically describe a very weak, bluish coloured flame which is barely visible (Kumar et al., 2002). The colour of the flame is also dependent on the type of fuel, with LPG fuel possibly resulting in a blue/green colour (Hasegawa et al., 1997), but this is dependent on the diluent (Cavaliere & De Joannon, 2004).

## 2. Application and implementation of MILD combustion

Driven by the industrial relevance, the MILD combustion process has so far predominately been implemented and investigated in furnaces. In most combustion systems, combustion efficiency is sufficiently high, but there is room for improvement in terms of heat utilization (Choi & Katsuki, 2001). Economical benefits are sought by increasing thermal efficiency, which can be accomplished by using preheating, and can achieve fuel savings up to 60% (Weber et al., 1999).

Large amounts of waste heat can be recovered in furnaces using heat-recovery devices to preheat the combustion air, typically above 1300 K (Katsuki & Hasegawa, 1998). To achieve preheating, energy from the exhaust gases is transferred back to the combustion air either by recuperative or regenerative heat exchangers (Plessing et al., 1998).

As an extension to using preheated air to achieve energy savings, dilution with exhaust gases has been seen to lower NO<sub>x</sub> emissions, although the low O<sub>2</sub> levels require even higher preheat temperatures than generally used for previous heat recovery systems (Choi & Katsuki, 2001). Using highly preheated air (> 1100 K), in conjunction with low O<sub>2</sub> levels (<15%), can accomplish energy savings, improved thermal performance and lower NO<sub>x</sub> emissions (Hasegawa et al., 1997). Worth highlighting is that although the oxygen levels used in MILD combustion are very low, on a global level, furnaces are operated lean (Kumar et al., 2002).

Preheating can also be achieved internal to the combustor by means of internal flow recirculation to provide heating of the unburnt gases (Kumar et al., 2002; Plessing et al., 1998). Due to the high temperatures, self-ignition in MILD combustion conditions is assured and so high-velocity jets can be used (Cavaliere & De Joannon, 2004). The use of high velocity jets is advantageous to ensure adequate mixing of the fuel, air and recirculated exhaust gases is achieved to simultaneously heat and dilute the reactants (Plessing et al., 1998). Such internal recirculation is the key principle of the FLOX burners which are at the forefront of the industrial application of MILD combustion (Wünning, J.A. & Wünning, J.J., 1997).

The key features of furnaces operating in the MILD combustion regime are the increased thermal efficiency, flat thermal field and lower NO<sub>x</sub> emissions. In relation to a conventional combustion chamber, the radiation flux is higher in the first zone on the combustor and lower in the second part, which overall leads to a more uniform thermal field (Cavaliere & De Joannon, 2004). In the work of Kumar et al. (2002), they found that the normalised spatial temperature variation is 15% for MILD, compared to 50% for their classical jet flame. Moreover, the heat flux uniformity achieved through MILD combustion is a highly desirable feature that often cannot be met with conventional burner technology (Hasegawa et al., 1997). The heat flux of a furnace operating in MILD regime is almost constant heat flux, which may provide high heat transfer rates, and is very desirable for a number of industrial processes (Weber et al., 2000). It is estimated that MILD combustion technologies are capable of a 30% energy saving (Katsuki & Hasegawa, 1998). Although achieving greater efficiency can be achieved using MILD combustion, improved product quality as a result of the thermal field is also a major driving force behind the process (Cavaliere & De Joannon, 2004).

The lack of flame and lower peak temperatures in MILD combustion may be thought to be less efficient at heat deposition, but because of the larger reaction volume the total heat radiation flux can actually be higher in MILD combustion than in conventional flames (Weber et al., 2000). This has been shown previously; where radiation emitted downstream from the reaction zone is about five times that emitted upstream (Williams, 1985), implying that although the peak temperature within MILD combustion is lower there is significant radiation emitted. Moreover, while chemiluminescence is low in MILD combustion, and may provide flames with their distinctive colours, the associated energy transfer by this mechanism is negligible in comparison with radiation from major stable compounds (such as H<sub>2</sub>O and CO<sub>2</sub>) (Williams, 1985). As an example of the increased efficiency offered by MILD combustion, Wünning, J.A. and Wünning, J.J. (1997) used CFD modelling to determine the input energy required to achieve 160 kW output from a furnace. Without any

preheating, the burner capacity was 400 kW, with 600 °C preheat using a recuperator the input was 245 kW (Wünning, J.A. & Wünning, J.J., 1997). For FLOX mode, with 950°C preheat the required input was as low as 200 kW – half that of the no preheat case (Wünning, J.A. & Wünning, J.J., 1997). Furthermore, the peak production rate of NO<sub>x</sub> is reduced by several orders of magnitude (Wünning, J.A. & Wünning, J.J., 1997). This highlights why industry is seeking this technology to deal with reducing fuel costs and meeting more stringent NO<sub>x</sub> emission targets (Brggraaf et al., 2005).

A schematic layout of a MILD combustion furnace is shown in Figure 2 from Choi & Katsuki (2001). This furnace is based on an alternating flow system, and comprises of two sets of regenerators. In one flow direction, the incoming air passes over a hot ceramic honeycomb, which preheats the incoming air. As the hot exhaust gases leave the furnace at the other end they pass through another ceramic honeycomb, and subsequently heat it. After a certain period of time, the temperature of the incoming honeycomb decreases, whilst that of the exhaust side honeycomb has increased, and the flow direction is then reversed. The now hot honeycomb acts to heat the incoming air, whilst the cooler honeycomb is now heated by the exhaust gases. The process is then again reversed and continues the alternating heating and cooling of the regenerators.

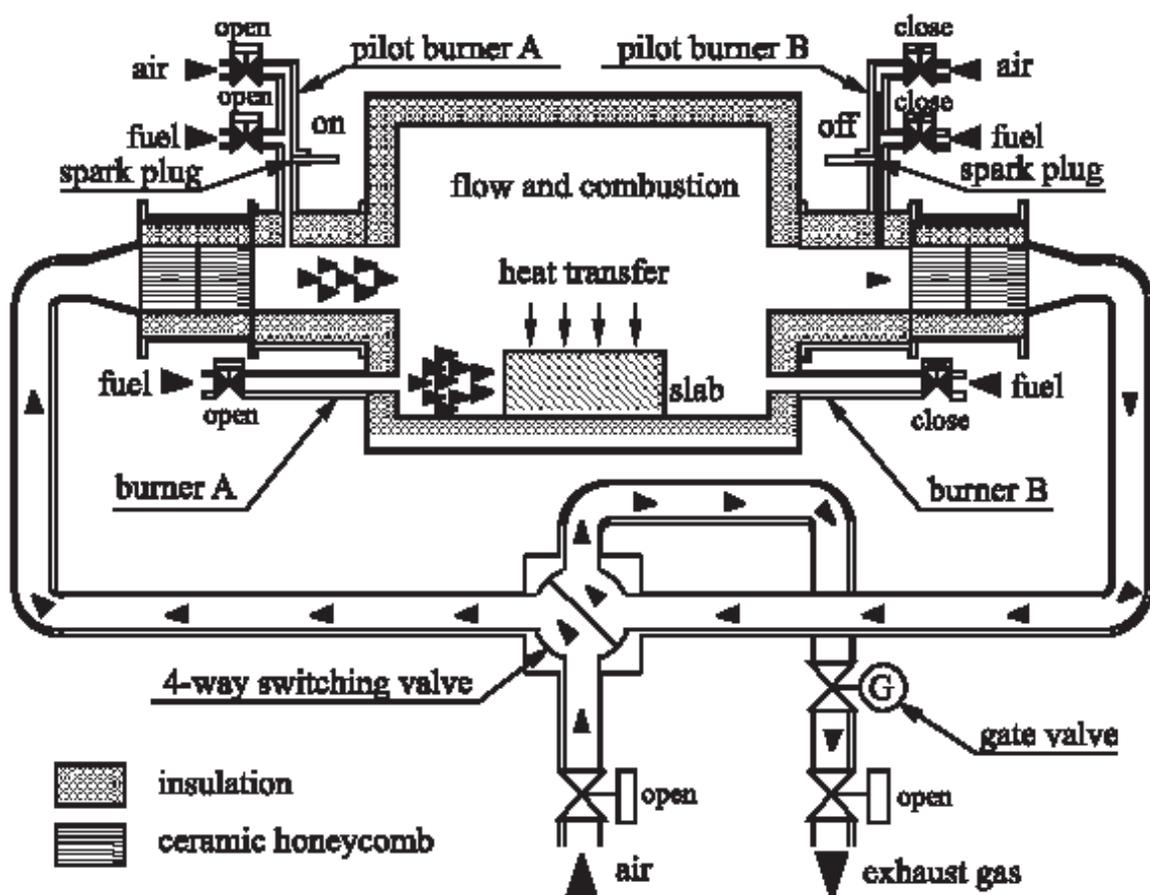


Fig. 2. Industrially implemented MILD combustion furnace (Choi & Katsuki, 2001)

In addition to furnace design, MILD combustion has potential for inclusion in other practical application too. The implementation used for furnaces clearly require significant alteration for use in other applications, but, at least in principle, the underlying aspects of

MILD combustion can be extended to other fields. The low temperature and uniform thermal fields are advantageous attributes in the design of gas turbines, particularly from the materials standpoint (Cavaliere & De Joannon, 2004). Advancement in the application of some aspects of MILD combustion to the design of gas turbines has been made, but issues with self-ignition and dilution has meant generalised application has not yet been achieved (Cavaliere & De Joannon, 2004).

Along a similar vein, some developments which are similar to MILD combustion have been applied to diesel engines but, to date these are not what could be considered a fully fledged MILD combustion system (Angrill et al., 2000; Choi & Katsuki, 2001). An engine which is much closer to practical implementation of MILD combustion is the Homogenous Charge Compression Ignition (HCCI) engine (Cavaliere & De Joannon, 2004).

The preheating of combustible mixtures, which is central to MILD combustion, offers the ability to use fuels that would not typically be used - in particular, low calorific value fuels (Choi & Katsuki, 2001; Katsuki & Hasegawa, 1998).

There is clearly potential for MILD combustion to offer significant benefits in a number of applications. One of the biggest impediments related to the implementation of this promising technology is the lack of fundamental knowledge of the reaction zone structure. A better understanding of the flame structure is needed to see widespread implementation of MILD combustion (Muruta et al., 2000).

### 3. MILD combustion studies

As was highlighted in the preceding section, MILD combustion has potential for use in a number of practical applications, but this is limited by a lack of detailed understanding of fundamental aspects. Although previous research has been conducted in the area of MILD combustion, this work has predominately concentrated on large scale systems. Despite the importance of these systems, such methodology fails to address the fundamental issues of the regime.

This project seeks to differ from much of the existing research by using a well controlled experimental burner. The use of such a burner enables a wide range of combustion parameters to be easily varied, whereas the existing research has been limited in this respect. In addition, the use of an experimental burner enables laser diagnostic measurement techniques to be employed, which cannot be used effectively in the existing systems. In this way, utilizing an experimental burner will avoid the limitations of previous investigations, whilst simultaneously enabling the required conditions to be emulated.

Although much of the previous study of MILD combustion has tended to be directed towards industrial systems, there have been some studies on the fundamental aspects. Due to the lack of visible flame, it has been necessary to resort to non-luminosity based techniques. As an example, Plessing et al. (1998) used laser induced predissociative fluorescence (LIPF) of OH to visualise the reaction zone. In conjunction with the OH-LIPF, Rayleigh measurements were also recorded. Rayleigh scattering is particularly suitable for MILD combustion systems as the high levels of dilution result in the Rayleigh cross-section varying by less than 2% between the burnt and unburnt gases (Plessing et al., 1998), further enhanced by the lack of particulate matter.

The furnace from Plessing et al. (1998), has been used in further studies, including Özdemir & Peters (2001), Coelho & Peters (2001) and Dally et al. (2004). Özdemir & Peters (2001) extended the OH and temperature measurements of the furnace by including flow field

measurements, finding that strong shear is an important criteria for attaining the high mixing rates necessary to create MILD combustion conditions. Coelho & Peters (2001) numerically simulated the same furnace and compared the results to the previous measurements. The models qualitatively matched the experimental results, although differences were noted near the burner exit and also in NO formation. The same furnace has also been used by Dally et al. (2004) to study the effects of fuel dilution, both numerically and experimentally. They found that the shift in stoichiometry caused by fuel dilution helped in the establishment of MILD combustion.

Other furnace designs have been studied both experimentally and numerically by Weber et al. (1999, 2000) and subsequently modelled by Mancini et al., (2002). Similar to Coelho & Peters (2001), Mancini et al. (2002) found that simulations matched the experiment finding, except near the jet.

On a more fundamental level, Dally et al. (2002a, 2002b) reported on the structure of hydrocarbon non-premixed laminar and turbulent flames stabilised on a jet in a hot and diluted co-flow (JHC) burner. They used single-point Raman-Rayleigh-LIF diagnostic techniques to simultaneously measure temperature, major and minor species at different locations in these flames. They found that major changes in the flame structure occur when reducing the oxygen concentration and that, at higher jet Reynolds number and low oxygen concentration, oxygen leakage from the surrounding may cause local extinction of the flame front.

As an extension to the experimental work of Dally et al. (2002a, 2002b), CFD modelling has subsequently been applied to the same flame conditions (Christo & Dally, 2005; Kim et al., 2005). In these CFD studies, the models have shown that the experimental results can be reproduced, albeit with significant deviations in some situations, furthermore, only some of the model variants gave good agreement. In all cases, obtaining reliable CFD models proved most difficult at the very low O<sub>2</sub> conditions, and at the downstream measurement locations where the entrainment of surrounding air introduces an additional degree of complexity. Overall, these modelling studies indicate that there is potential for application of CFD to MILD combustion conditions, but some further work is still required.

In a study of a laboratory MILD combustion burner it has been shown that reactions under the MILD combustion regime occur in disconnected zones where the OH and temperature intensities differ (Dally & Peters, 2002; Plessing et al., 1998).

The explanation for this discontinuity between the reaction and the temperature is attributed to the varying amounts of recirculated exhaust gases, which change the balance required for MILD combustion. The conclusion of the work of Plessing et al. (1998) is that MILD combustion resembles a reaction in a well-stirred reactor, although further investigations are necessary for complete analysis of MILD combustion.

It has been suggested that a furnace operating under MILD conditions is similar to a well-stirred reactor (WSR) (Weber et al., 1999). Based on the concept of attempting to infer MILD combustion from a well-stirred reactor, De Joannon et al. (2000), has attempted to model a WSR with MILD combustion conditions. This work did not compare the composition of a well-stirred reactor to any experimental data, but did examine the effect of the diluent composition. They found that there is a dramatic effect on the combustion composition, depending on the composition of the diluent. Also the effect of the residence time on the reaction was of no significance for the majority of the temperature range 500 – 2000 K. However as identified in their work, De Joannon et al. (2000), acknowledge that a well-stirred reactor is unfeasible for a practical combustor.

#### 4. The potentialities of the MILD combustion in application related to the pollutant emission

Several works exploit the influence of MILD combustion on formation of  $\text{NO}_x$  through the different reaction paths. It is well known that nitrogen oxides form along there possible paths, namely thermal  $\text{NO}_x$ , fuel- $\text{NO}_x$  and prompt  $\text{NO}_x$ . The first two mechanisms, which are more efficient in the  $\text{NO}_x$  formation, are depressed by the low-temperature in MILD combustion. The first mechanism is most effective for high levels of temperature and oxygen, according to the Zel'dovich mechanism, whereas the second one is maximized for high levels of fuel richness and fuel nitrogen content. The last mechanism relies on high reactant temperature also and can proceed in very rich conditions only if the temperature is high enough to sustain the process.

Wünning, J.A. & Wünning, J.J. (1997) reviewed flameless oxidation using experimental and numerical techniques. The study reveals the ability of flameless combustion to lower  $\text{NO}_x$  emissions through lowered adiabatic flame temperatures. The study, however, used low inlet air temperatures and did not include a reaction flow analysis study.

Ju & Niioka (1997) studied  $\text{NO}$  formation in a non-premixed two-dimensional laminar jet flame, another canonical configuration for the understanding of the effects of turbulence on combustion. The study shows that the formation of prompt  $\text{NO}$  is observed in the flame reaction zone, while the thermal  $\text{NO}$  mechanism is the predominant path for  $\text{NO}$  formation on the high-temperature air side. The emission index of  $\text{NO}$  was shown to have a high sensitivity to the oxidizer preheat temperature, increasing drastically with an increasing preheat temperature.

Hamdi et al. (2004) investigated the high temperature air combustion of partially premixed (lean or rich) flames (methane/air) in counter-flow geometry (figure 3). The study illustrated that the flame structure for the rich partially premixed flame is significantly affected by the oxygen concentration in the air and a significant reduction in  $\text{NO}_x$  emission is observed. However,

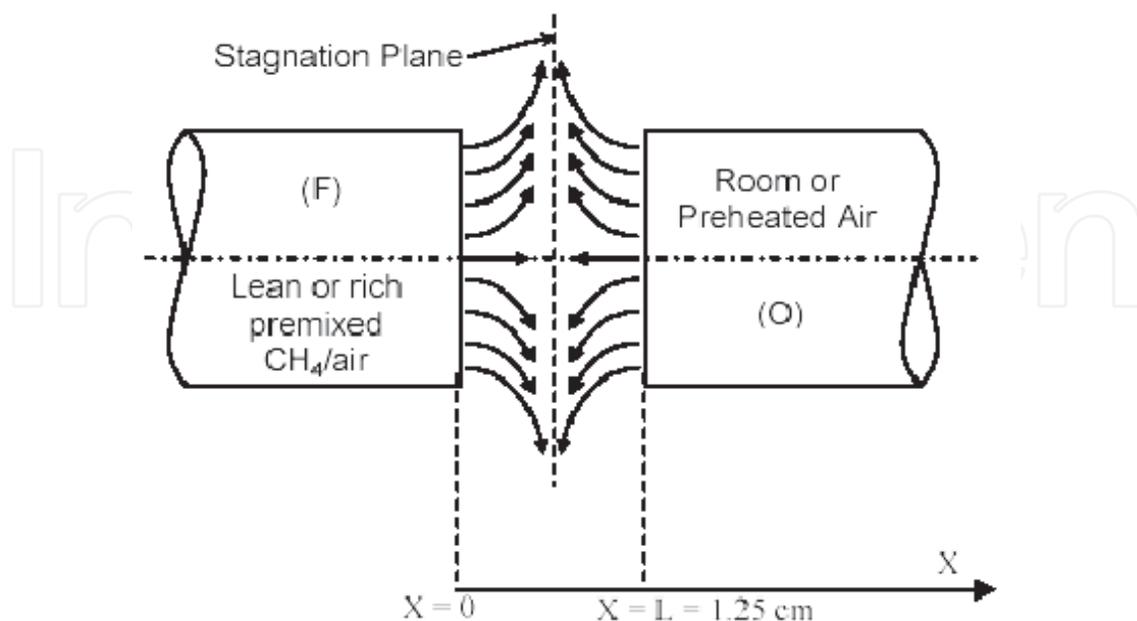


Fig. 3. Opposed jet flame schematic; Preheat air vs. Lean or rich premixed methane/air (Hamdi et al., 2004)

Figure 4 reports several accumulated  $\text{NO}_x$  data in a log scale as a function of process temperature assuming very efficient preheating of the combustion air (60-80% of the process temperature).

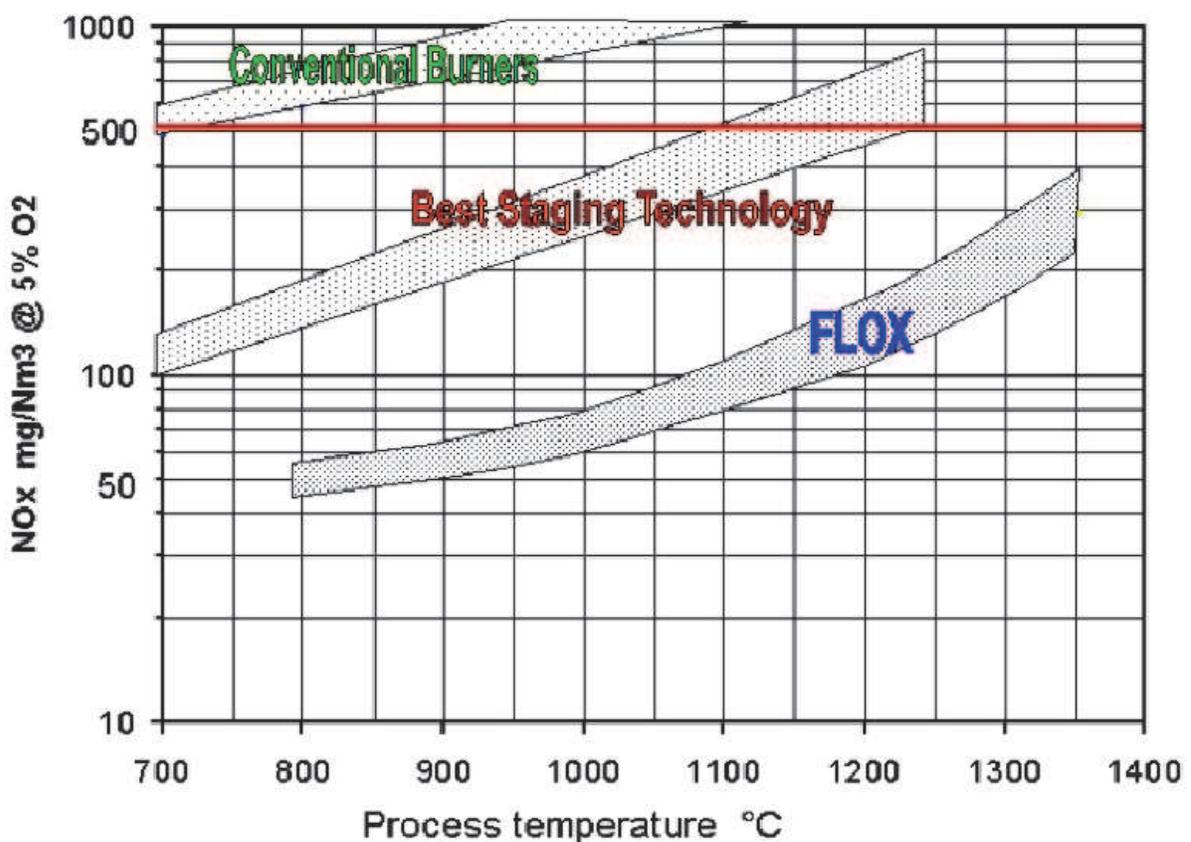


Fig. 4.  $\text{NO}_x$  emissions vs. process temperature (Milani & Saponaro, 2001)

It may be seen from Figure 4 that MILD combustion (region labelled FLOX) may abate  $\text{NO}_x$  emissions by one order of magnitude even with respect to the best staging techniques for Low- $\text{NO}_x$ , envisaged for natural gas firing. The main reason for this excellent result stem from the well known circumstance, that thermal NO formation is extremely sensitive to flame temperature peaks or spikes and these are now cut away in MILD combustion firing. But also the other known NO formation mechanisms are positively modified, as prompt NO depends on radicals that are abundant in a flame front, but much reduced in MILD combustion mode (Plessing et al., 1998) and also fuel NO may undergo reburning effects capable of reconvertng NO into  $\text{N}_2$  species (Eddings & Sarofin, 2000; Weber et al., 2000).

Recently, Hamdi et al. (2009) studied the effects of fuel dilution and strain rate on zone structure and  $\text{NO}_x$  formation in MILD combustion. The study is based on parametric simulations of non-premixed counter-flow flames of methane with nitrogen dilution and highly preheated air.

A counter-flow laminar flame, therefore, is the best field if we want to discuss the local combustion characteristics of the industrial furnace, in particular, using detailed chemical

kinetics. Although the real counter-flow flame is inherently two-dimensional, the assumption and formulation of a similarity solution (Giovangigli & Smooke, 1987; Kee et al., 1987; Darabiha et al., 1988) are applied to transform the problem into one-dimensional. Therefore, one-dimensional solutions along  $x$  axis represent the whole field of the flat flame formed perpendicular to  $x$  axis by the similarity. The governing equations of the flow can be found in refs. (Giovangigli & Smooke, 1987; Kee et al., 1987; Darabiha et al., 1988).

The solution is computed using Sandia's OPPDIF code (Lutz et al., 1996) along with the GRI-Mech 3.0 mechanism (Glassman, 1996) for methane and  $\text{NO}_x$  chemistries.

Through their study, it can be seen in figure 5, that there is a general decrease in the production of  $\text{NO}$  and  $\text{H}_2$  with a decrease in fuel concentration. This is due to the reduced fuel present to transport to the flame.

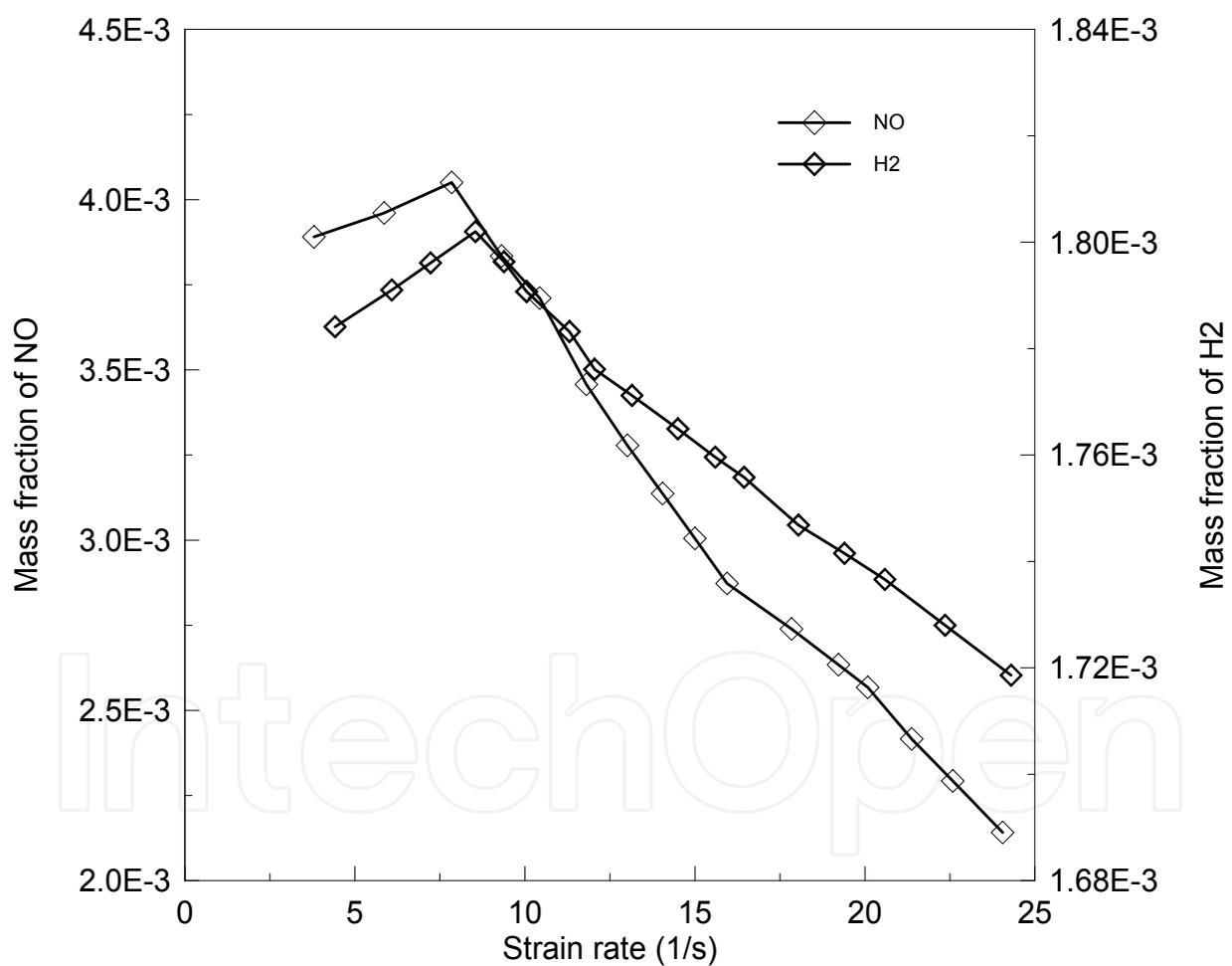


Fig. 5. Mass fractions for  $\text{NO}$  and  $\text{H}_2$  vs. strain rate at air preheated temperature of 2000K and fuel concentration of 15%

The emission index of  $\text{NO}$  was found to decrease as fuel concentration is lowered due in part to the weakening reaction as fuel concentration is decreased, as shown in figure 6.

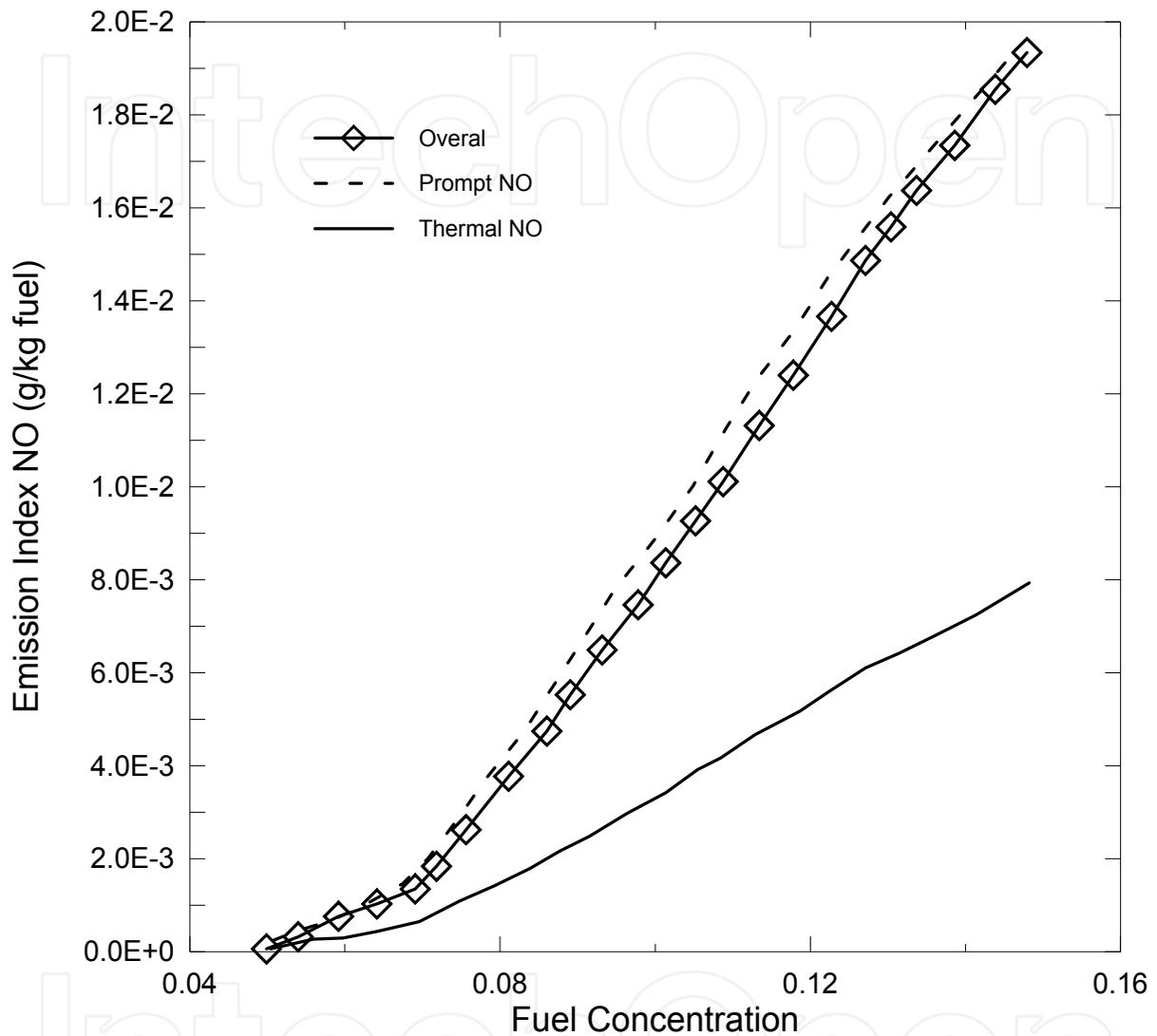


Fig. 6. Emission index profiles for  $\text{NO}_x$  vs. fuel concentration at air preheated temperature of 1000K and strain rate of  $4\text{s}^{-1}$

While there is a decrease in the overall contribution of both thermal and prompt NO as fuel concentration is decreased, there is also a decrease in the individual contributions of thermal and prompt NO, as indicated in figure 7. Due to the high dependence on temperature seen in the thermal NO mechanism, the thermal NO mechanism is dominant at higher preheat temperatures, while the prompt is dominant at lower preheat temperatures.

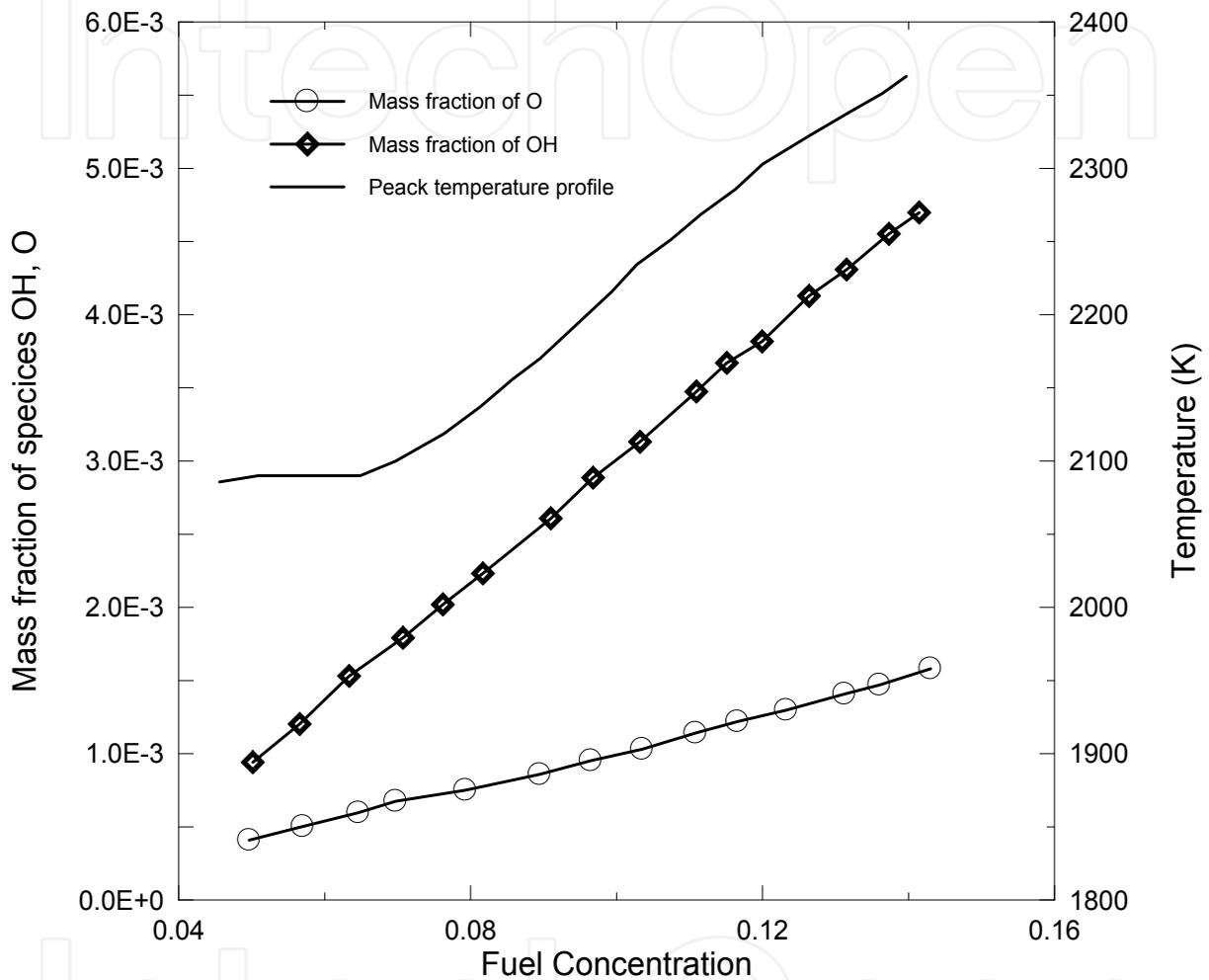


Fig. 7. Mass fractions of O and OH vs. fuel concentration at air preheated temperature of 2000 K and strain rate of  $24 \text{ s}^{-1}$

Hamdi et al. (2009) found, in contrast with fuel concentration, that strain rate has been seen to have a large effect on the slower mechanisms, but not the fast ones due to a balance between decreased residence times and higher transport and vice versa, as illustrated by figure 8.

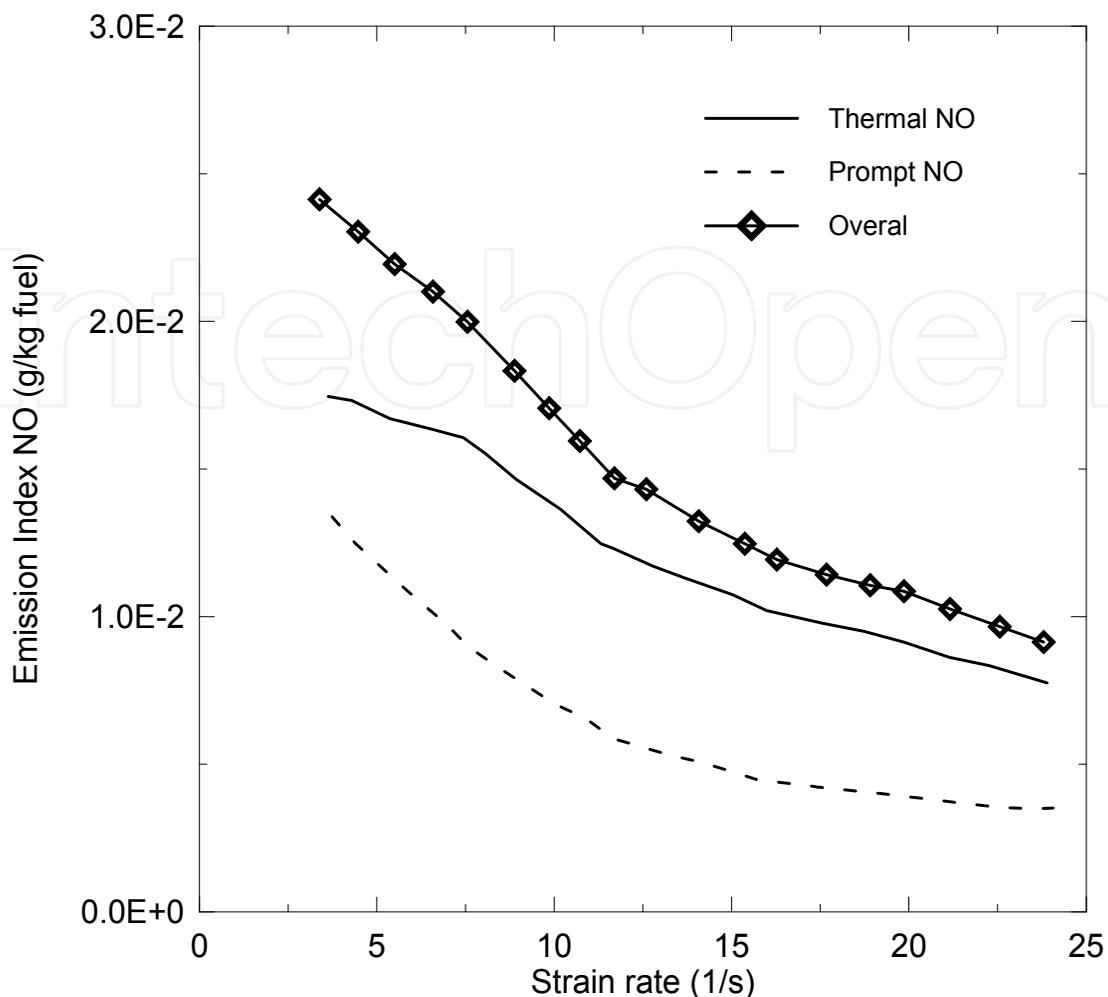


Fig. 8. Emission index profiles for  $\text{NO}_x$  vs. strain rate at air preheated temperature of 2000 K and fuel concentration 5%

### 5. Turbulence-chemistry interaction in MILD combustion

Modeling the interaction of a complex set of chemical reactions and high Reynolds number turbulent fluid flow typical for gas turbine combustion is a challenging task. If the emphasis is on the formation of pollutants such as  $\text{NO}_x$  a full chemistry model is prohibitive. Direct numerical simulation is not feasible due to high computational costs and storage requirements. Even a probability density function (PDF) modeling approach of a "real" gas turbine combustion chamber with boundary effects, swirling inhomogeneous flow by far exceeds computational capacities. Therefore, severe simplifications have to be made to be able to study the interaction of turbulence and detailed chemistry with respect of  $\text{NO}_x$  formation. In the MILD combustion model, the combustor was simulated by the flow model shown in figure 9; the hot gases exit the first combustor chamber and enter the second combustor chamber.

The first combustion chamber is assumed to be an ideal, turbulent, adiabatic, constant pressure, well stirred reactor (PSR). It is assumed there are no boundary effects and the turbulence created at the inlet is homogeneous, isotropic, and stationary. Then, the second combustion chamber is described as a "Partially Stirred Reactor (PaSR)", where mixing and

chemical reactions occur simultaneously. The widely available CHEMKIN package (Miller et al., 1996) and specially its software application PaSR is used to model the flow field that occur in the 2<sup>nd</sup> combustion chamber.

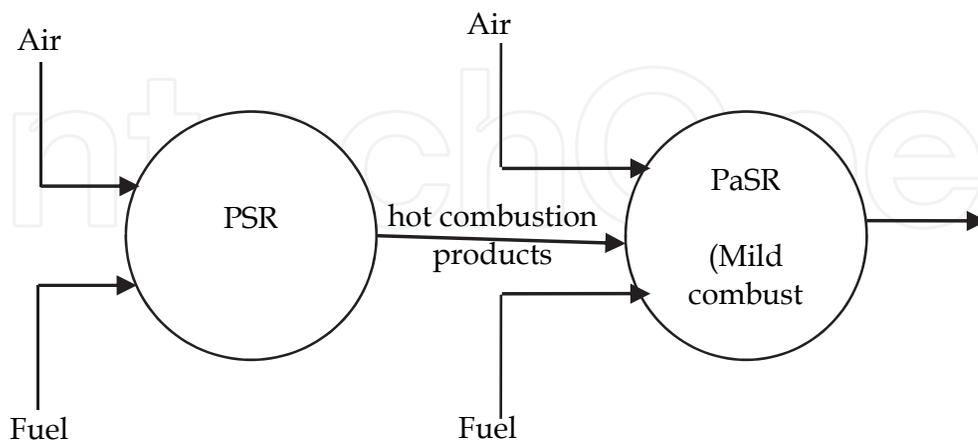


Fig. 9. Principle of the MILD combustion model

### 5.1 Reactor equations

The Partially Stirred Reactor (PaSR) is an extension of a Continuously Stirred Tank Reactor (CSTR) model that addresses the interaction between chemical reactions and turbulence (Chen, 1997; Correa, 1993). The basic assumptions for the PaSR are similar to those of the CSTR or PSR (perfectly Stirred Reactor). The major difference between a PSR and a PaSR lies in the treatment of the molecular mixing inside the reactor. In a PSR the contents of the reactor are well mixed by assuming high-intensity turbulent stirring action and the only influence from fluid dynamics is controlled by the reactor residence time  $\tau_R$ . Unlike the PSR, a PaSR allows fluid dynamics to control the extent of the mixing and consequently the chemical reactions by means of an additional parameter: the scalar mixing time,  $\tau_{\text{mix}}$ . The turbulent mixing time scale is often considered to be proportional to the turbulent eddy turnover time as (Kee et al., 2002):

$$\tau_{\text{mix}} = C_D \frac{\tilde{k}}{\tilde{\epsilon}} \quad (2)$$

Where  $C_D$  usually treated as a constant but its value varies for different flow configurations, as suggested by Pope (1991).  $C_D$  is set  $C_D = 2.0$ . The ratio of turbulent kinetic energy to its dissipation rate,  $\tilde{k}/\tilde{\epsilon}$ , represents the time scale of the energy-containing eddies which characterize the turbulent mixing action.

The composition and temperature in the PaSR are described by a probability density function (PDF). This composition PDF is a subset of the joint velocity-composition PDF because the flow field in the PaSR is assumed to be spatially homogeneous. Velocity fluctuations are also ignored; that is, the PDF is over scalars only, but is not a delta-function

in scalar space because reactants, intermediates, and products are not mixed at the molecular level.

The PaSR consists of an adiabatic chamber having  $M$  inlet streams and one outlet. Steady flows of reactants are introduced through the inlets with given gas compositions and temperatures. The reactor pressure is assumed to be constant.

The overall mass balance for the gas mixture inside the PaSR is:

$$\frac{d\langle \rho \rangle V}{dt} = \sum_{i=1}^M \dot{m}_i - \dot{m}_0 = 0 \quad (3)$$

where  $\dot{m}_i$  is the mass flow rate of the  $i^{\text{th}}$  inlet and  $\dot{m}_0$  is the through-flow mass flow rate.

The average properties of the PaSR are obtained from the ensemble of particles inside the reactor. Each particle is regarded as an independent PSR and interacts with others only through the molecular mixing process. Therefore, the conservation of energy and species is applied to an individual particle rather than to the reactor.

The species equation for a particle is then similar to that of a PSR:

$$\frac{dY_k^{(n)}}{dt} = \frac{1}{\dot{m}_0 \tau_R} \sum_{i=1}^M \left\{ \dot{m}_i (Y_{i,k} - \langle Y_k \rangle) \right\} + \frac{W_k \omega_k^{(n)}}{\rho^{(n)}} \quad (4)$$

and so is the energy equation for a particle is:

$$\frac{dT^{(n)}}{dt} = \frac{1}{C_p^{(n)} \dot{m}_0 \tau_R} \sum_{i=1}^M \dot{m}_i \left( \sum_{k=1}^{k_g} Y_{i,k} (h_{i,k} - \langle h_k \rangle) \right) - \sum_{k=1}^{k_g} \frac{W_k \omega_k^{(n)} h_k^{(n)}}{\rho^{(n)} C_p^{(n)}} \quad (5)$$

In the above equations, the angled bracket ( $\langle \rangle$ ) indicates the ensemble average that we use to approximate the density-weighted average in the simulation. The average residence time of the reactor,  $\tau_R$ , is calculated as:

$$\tau_R = \frac{\langle \rho \rangle V}{\dot{m}_0} \quad (6)$$

In the following section we discuss the influence of the turbulent mixing on the chemical kinetics under MILD combustion conditions. In order to demonstrate the influence of turbulent mixing on the chemical reactions we vary the turbulent mixing time  $\tau_{\text{mix}}$ . From the limiting case of the PSR (i.e.  $\tau_{\text{mix}} = 0$ s), which corresponds to very fast mixing, we move to realistic turbulent time scales  $\tau_{\text{mix}} = 50$  ms, which is slow mixing and an intermediate mixing time  $\tau_{\text{mix}} = 25$  ms.

The computational results show, through figure 10, that the combustion processes as well as the  $\text{NO}_x$  formations are very sensitive on the mixing intensity. With increasing turbulent mixing time (i.e. decreasing mixing intensity) the combustion process is stretched out. The ignition delay is shorter but the residence time to achieve complete combustion increase significantly.

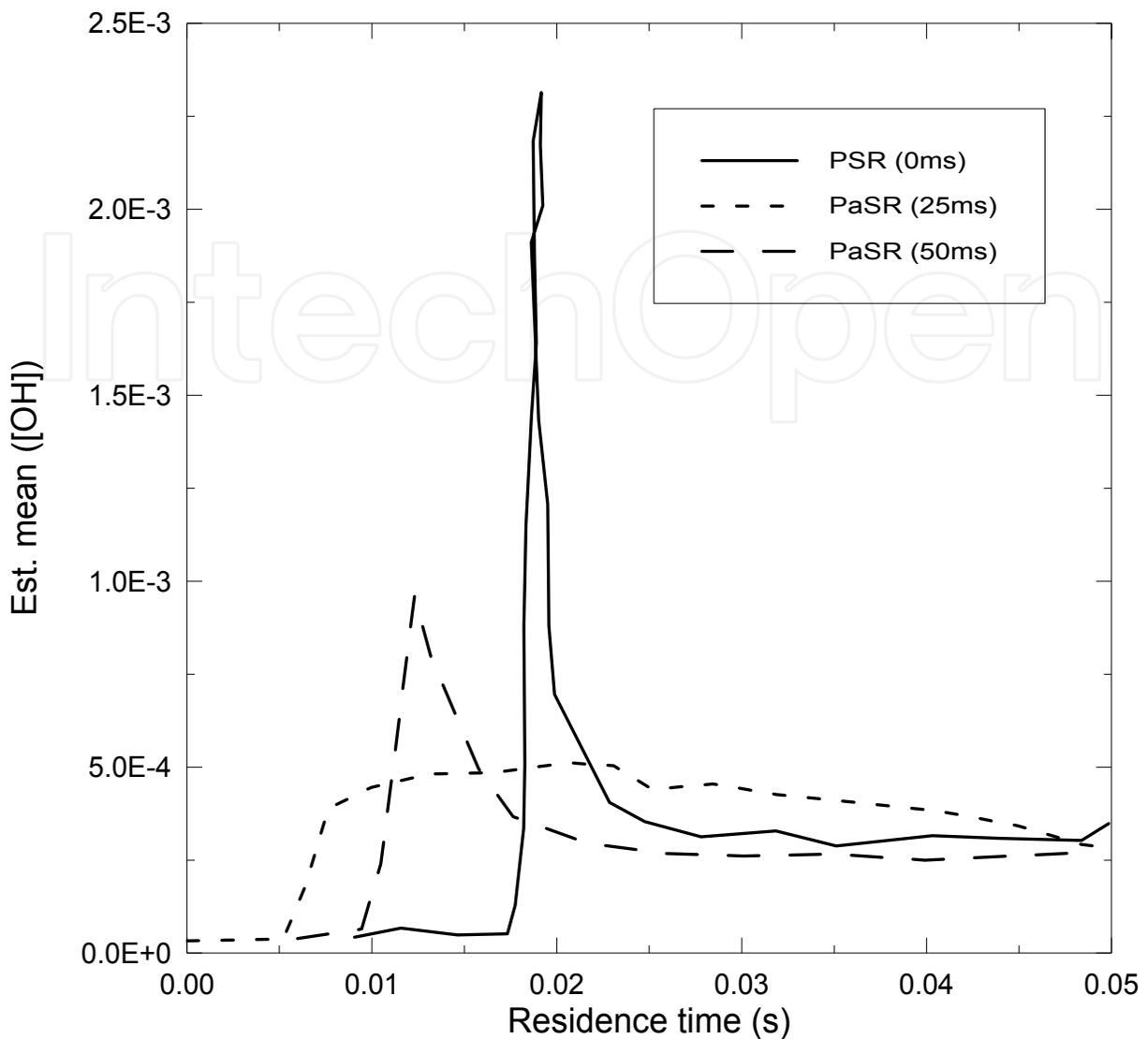


Fig. 10. Time evolution of estimated mean of OH mass fraction in the PSR and PaSR cases

## 6. Conclusions

This review chapter collects information which could be useful in understanding the fundamentals and applications of MILD combustion. The pieces of information in this field are still sparse, because of the recent identification of the process, so that many speculative considerations have been presented in order to make the whole framework more consistent and rich with potential new applications.

The main points to be stressed as concluding remarks pertain to three main considerations. The first is that MILD Combustion has to be considered a new combustion regime. It is a combustion process that achieved a desirable combination of low pollutant emissions and improved fuel savings.

The second consideration is that MILD combustion can find a possible application in fuel reforming to produce hydrogen. However the three parameters (fuel concentration, strain rate and oxidizer preheat temperature) discussed in the study described in Section 4, play major roles in the balance between the desired product (hydrogen) and the undesired by-product (NO).

The third consideration is that the characteristic times of kinetics and turbulence, for the MILD combustion regime, become comparable and the two phenomena are coupled with each other. It is therefore of great interest to determine the interaction of the turbulent mixing and the combustion process as well as its impact on NO<sub>x</sub> emission. This study has been described in Section 5. A Partially Stirred Reactor model has been developed to study the influence of turbulent mixing intensity on MILD combustion and NO<sub>x</sub> formation in gas turbine combustor.

### Nomenclature

$C_D$	Model constant (= 2.0)
$C_P$	Specific heat at constant pressure [J/kg K]
$h_k$	Specific enthalpy of species k [J/kg]
$K_V$	Recirculation rate [-]
$\tilde{k}$	Turbulent kinetic energy [m <sup>2</sup> /s <sup>2</sup> ]
$k_g$	Total of species included in the gas-phase
$\dot{M}$	Mass flow rate [kg/s]
$\dot{m}_0$	Through-flow mass flow rate [kg/s]
$\dot{m}_i$	Mass flow rate of the <i>i</i> <sup>th</sup> inlet [kg/s]
$T$	Temperature [K]
$t$	Time [s]
$V$	Reactor volume [m <sup>3</sup> ]
$W_k$	Molecular weight of species k [kg/mol]
$Y_k$	Mass fraction of species k [-]

### Greek scripts

$\tilde{\epsilon}$	Dissipation ratio of turbulent energy [m <sup>2</sup> /s <sup>3</sup> ]
$\rho$	Density [kg/m <sup>3</sup> ]
$\tau_{mix}$	Turbulent mixing time [s]
$\tau_R$	Reactor residence time [s]
$\omega_k$	Reaction rate of species k [s <sup>-1</sup> ]

### Subscripts

A	Clean combustion air
E	Recirculated exhaust gas
F	Fuel
<i>i</i>	<i>i</i> <sup>th</sup> inlet
<i>k</i>	<i>k</i> <sup>th</sup> species
O	Oxidizer

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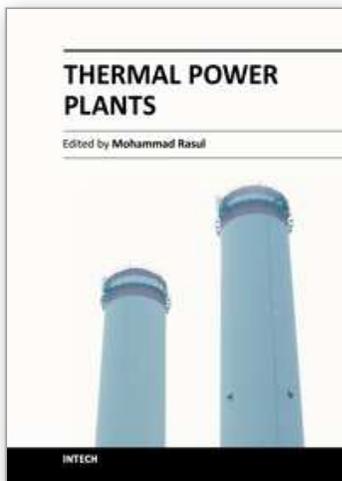
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