Thermodynamic aspects and heat transfer characteristics of HiTAC furnaces with regenerators

Nabil Elias Rafidi

Doctoral Dissertation

Royal Institute of Technology
School of Industrial Engineering and Management
Department Of Materials Science and Engineering
Division of Energy and Furnace Technology
Se- 100 44 Stockholm
Sweden
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Abstract

Oxygen-diluted Combustion (OdC) technology has evolved from the concept of Excess Enthalpy Combustion and is characterized by reactants of low oxygen concentration and high temperature. Recent advances in this technology have demonstrated significant energy savings, high and uniform thermal field, low pollution, and the possibility for downsizing the equipment for a range of furnace applications. Moreover, the technology has shown promise for wider applications in various processes and power industries.

The objectives of this thesis are to analyze the thermodynamic aspects of this novel combustion technology and to quantify the enhancement in efficiency and heat transfer inside a furnace in order to explore the potentials for reduced thermodynamic irreversibility of a combustion process and reduced energy consumption in an industrial furnace. Therefore, theoretical and experimental investigations were carried out.

The 2nd law of thermodynamics analyses of OdC systems have been carried out for cases in which the oxidizer is either oxygen (Flameless-oxy-fuel) or air (High Temperature Air Combustion, HiTAC). The analyses demonstrate the possibilities of reducing thermodynamic irreversibility of combustion by considering an oxygen-diluted combustion process that utilizes both gas- and/or heat-recirculation. Furthermore, the results showed that an oxygen-diluted combustion system that utilizes oxygen as an oxidizer, in place of air, results in higher 1st and 2nd law efficiencies.

Mathematical models for heat regenerators were developed to be designing tools for maximized heat recovery. These models were verified by heat performance experiments carried out on various heat regenerators.

Furthermore, experiments were performed in a semi-industrial test furnace. It was equipped with various regenerative burning systems to establish combustion and heat transfer conditions prevailing in an industrial furnace operating based on HiTAC. The tests were carried out at seven firing configurations, two conventional and five HiTAC configurations, for direct and indirect heating systems.

Measurements of energy balance were performed on the test furnace at various configurations in order to obtain the 1st law efficiency. Moreover, local measurements of temperature, gas composition, and heat fluxes in the semi-industrial test furnace were performed to find out the main characteristics of HiTAC flame and the effects of these characteristics on the heating potential, i.e., useful heating in the furnace. In the case of HiTAC, these measurements showed uniformities of chemistry, temperature, temperature fluctuation, and heat fluxes profiles. The values of fluctuations in temperature were small. The high speed jets of the fuel and air penetrated deep into the furnace. The fuel gradually disappeared while intermediate species gradually appeared in relatively high concentrations and at broader regions inside the furnace. These findings indicate: a large reaction zone, low specific combustion intensity in the flame, low specific fuel energy release, and high heat release from this large flame. In addition to the thermodynamic limitations to the maximum temperature of the Oxygen-diluted Combustion, the low specific energy release of the fuel and the high heat release from the flame to its surroundings cause this uniform and relatively moderate temperature profile in a HiTAC flame, consequently suppressing thermal-NO formation.

Heat flux and energy balance measurements showed that heating potential is significantly increased in the case of HiTAC compared to that in the conventional case, implying much more energy savings than the apparent heat recovery from the heat regenerators, and consequently much less pollutants emissions. Therefore, it is certain that this large HiTAC flame emits more thermal
radiation to its surroundings than the conventional flame does, in spite of the moderate-uniform temperature profile of the flame. This intense heat flux was more uniform in all HiTAC configurations, including the indirect heating configuration, than that of the conventional-air combustion configuration.

**Keywords**

Combustion, flameless, oxy-fuel, heat-recirculation, gas-recirculation, heat transfer, furnace, radiant-tube, regenerative burner, honeycomb, fixed-bed.
Acknowledgements

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I would like to thank my supervisor, Professor Włodzimierz Blasiak, head of the division of Energy and Furnace Technology, for giving me the opportunity to become a team member in the above mentioned project and introducing me to the new promising combustion field, HiTAC, flameless, and etc. He was the driving force in my work; this work would not have been possible without his constant guidance, support, and encouragement.

I wish to express sincere appreciation to all my colleagues at the division of Energy and Furnace Technology: Alberto Tsamba, Anna Ponzo, Carlos Lucas, Dr. Darek Szewczyk, Jonas Broback, Krishna Narayanan, Magnus Mörtberg, Patrik Wikström, Dr. Reza Fakhrai, Ruchira Abeyweera, Dr. Simon Lille, Sivalingam Senthoor, Dr. Sylwester Kalisz, Prof. Tomasz Dobski, Dr. Wiehong Yang, and to all who came as visiting scientists or students, for their great help throughout the work and for the scientific and cultural exchanges.

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Finally, but most importantly, to my love and wife, Hanan: I owe you a debt of gratitude for your unlimited patience, encouragement, and understanding.
# List of Supplements

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<td>Rafidi N., Blasiak W., Thermodynamic aspects of oxygen-deficient combustion, Archives of Thermodynamics, ISSN 1231-0956, Vol. 26(2005), No. 2, 29-44.</td>
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List of Other Papers

The following publications are partly included in the thesis:


2- Rafidi N., Blasiak W., Simulation of fixed bed regenerative heat exchangers used in high temperature air combustion (HiTAC) burners, Applied Thermal Engineering, Article submitted, 2005.

3- Rafidi N., Blasiak W., Ceramic regenerative heat exchangers used in high temperature air combustion (HiTAC) burning system, Proceedings of Eighth International Conference on energy for a clean environment, Clean Air Conference in Lisbon, Portugal, 27-30 June 2005.


Patent:

Title: ‘A Method and a Meter for Measuring Heat Transfer Parameters in a Hot Environment’ by which the radiative heat flux, the convective heat transfer coefficient and gas temperature are measured at once in a boiler or a furnace. Inventor(s): Nabil Rafidi, Swedish patent application filing date: 05/08/2004, file #: 0 -401979- 0.
To my lovely family

Hanan

and

Elias
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Nomenclature

$A$  
area (m$^2$)

$a_i$  
specific heat transfer area (m$^2$/m$^3$)

$A_c$  
free-flow Area (m$^2$)

$B$  
matrix wall thickness (m)

$BC$  
boundary condition

$C$  
flow-stream capacity rate (W/K)

$C_r$  
matrix capacity rate (W/K)

$c_p$  
heat capacity (kJ/kg.K)

$\bar{c}_p$  
mean molar isobaric exergy capacity

$cs$  
specific heat of solid (kJ/kg.K)

$dp$  
particle diameter

$D_h$  
hydraulic diameter $[4.A_c/P_m]$ (m)

$E_{sys}$  
stored energy in a system (kJ)

$\dot{E}$  
exergy rate (kW)

$ERR$  
energy recovery rate (%)  

$FSR$  
flue gas suction ratio (%) 

$G$  
mass flux of a gas (kg/m$^2$ s)

$g$  
acceleration of gravity (m/s$^2$)

$Gr$  
thermal radiation incidence (kW/m$^2$)

$h$  
heat transfer coefficient (W/m$^2$K), specific enthalpy (kJ/kg)

$\dot{h}$  
irreversibility rate (kW)

$k$  
thermal conductivity (W/m.K)

$L$  
length of regenerator (m)

$M$  
molecular weight

$m$  
mass (kg)

$m$  
mass flow rate (kg/s)

$n$  
stoichiometric coefficient, number of moles

$p$  
pressure (pa)

$P_m$  
perimeter (m)

$q$  
heat flux, (kW/m$^2$)

$\dot{Q}$  
physical/chemical enthalpy rate (kW)

$\dot{Q}_{sys}$  
rate of heat added to a system (kW)

$\dot{Q}_{sur}$  
rate of surface heat loss (kW)

$R$  
flue gas recirculation ratio, universal gas constant

$R_f$  
flame occupation coefficient

$R_{HF}$  
heat flux ratio

$R_o$  
oxidation mixture ratio

$R_U$  
spatial uniformity ratio

$S$  
absolute entropy rate (J/K.s)

$\bar{T}$  
temperature ($^\circ$C) or ($^\circ$K)

$t$  
time (s)

$t_s$  
switching time (s)

$U$  
measured parameter

$U$  
velocity (m/s)

$V$  
flow rate (Nm$^3$/h)

$\dot{W}_{sys}$  
rate of work done by the system (kW)

$x, y, z$  
Cartesian coordinates

$x$  
molar fraction

$Z$  
elevation (m)

Greek symbols

$\alpha$  
surface absorptivity

$\Delta$  
differential

$\delta$  
efficiency defect

$\varepsilon$  
efficiency

$\bar{\varepsilon}$  
specific molar exergy (kJ/kmol)

$\eta$  
efficiency

$\eta_T$  
cold-side temperature efficiency

$\mu$  
viscosity

$\rho$  
density

$\sigma$  
Stefan-Boltzmann constant (W/m$^2$K$^4$)

$\tau$  
time constant (seconds)

$\phi$  
porosity

$\psi$  
$2^{nd}$ law efficiency

$\bar{\sigma}$  
mass fraction

$\varepsilon$  
surface emissivity

Dimensionless Groupings

$Bi$  
Biot number $((b/2) \ h \ /k_s)$  

$Pr$  
Prandtl number $(\mu C_p/k)$

$NTU$  
number of heat transfer units

$Nu$  
Nusselt number $\left(hD_h/k_s\right)$

$Re$  
Reynolds number $(\rho D_h u/\mu)$

$St$  
Stanton number $(Nu/R.Pr)$
Subscripts and Superscripts

<table>
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<tr>
<th>Subscript/Superscript</th>
<th>Description</th>
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<td>average</td>
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<tr>
<td>$b$</td>
<td>bulk</td>
</tr>
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<td>convective, cooling air</td>
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<tr>
<td>$ch$</td>
<td>chemical</td>
</tr>
<tr>
<td>$cm$</td>
<td>compensated value for time constant</td>
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<td>$comb$</td>
<td>combustor</td>
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<tr>
<td>$s$</td>
<td>solid, surface</td>
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<td>$w$</td>
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1. Background

The two main objectives in the development of combustion technology are to decrease energy consumption and to reduce pollutant emissions, e.g., NOx and CO2. One of the most widely used measures to reach ultimate thermal efficiency in furnaces is waste heat recovery, e.g., heat recirculation combustion. Heat recirculation combustion is a fuel saving combustion that is achieved by efficient recovery of waste heat in exhaust gases and uses this heat in pre-heating combustion air, i.e., the oxidizer. Pre-heated air adds enthalpy to the combustion zone, and therefore the term “Excess Enthalpy Combustion” was coined by Weinberg [1]. However, all temperature profiles inside a combustion chamber will rise up to some extent as a result of this method; see the high temperature flame area in Figure 1. This increase in flame temperature results in undesirable effects, e.g. rapid formation of nitric oxides emission (NOx), and overheating of materials used in furnaces. Nonetheless, the Excess Enthalpy Combustion shed new light on the next generation of advanced energy conversion technologies in the past decade.

In fact, an effective method for reducing flame temperature, and thereby NOx formation, is the exhaust gas-recirculation, i.e., the mixing of flue gases and combustibles. However, if ambient or cold combustion air, i.e., the oxidizer, is mixed and diluted with flue gases, oxygen levels in the reactants are reduced and combustion will mostly not happen, as in the non-combustible area in Figure 1. On the other hand, reactants, whose oxygen concentration is as low as 5%, can sustain combustion when pre-heated over the auto-ignition temperature; see the new combustion area in Figure 1. In fact, this regime of combustion is what concerns this entire thesis. This novel combustion technology couples the high thermodynamic efficiency with other unique features and characteristics including low NOx formation. It came in the middle of the 1990s with parallel research efforts made mainly in Japan and Germany.

Combustion that is characterized by reactants of low oxygen content and high temperature, does not only achieve reduction in energy consumption and pollutant emissions but also provides temperature and heat transfer uniformity in the combustion chamber, thereby contributing to enhancing the product quality in most industrial applications, downsizing the physical size of the combustion chamber, and/or increasing the production rate [2]. Moreover, a wide range of fuels, including the low calorific value and industrial gases can be utilized in this technology.

In literature many terms were used to refer to this technology and most of these terms are described in the review paper of Cavaliere et al. [3]. They refer to it as ‘Mild combustion’ because the temperature increase due to the reaction is lower or milder than the temperature of the pre-heated reactants.

Figure 1. Combustion regimes as a function of pre-heat temperature and oxygen content.
However, the most common term used by many researchers, including me and my colleagues at KTH, is \textit{High Temperature Air Combustion} (HiTAC). Flameless combustion and high temperature combustion technology (HiCOT) are a few other examples. In fact, there is a distinction between flameless and HiTAC in which HiTAC is characterised by high efficiency heat recovery. This distinction was made by many burner manufacturers who innovated the use of compact honeycomb regenerative heat exchangers.

Further innovations of this combustion phenomenon were continued, but instead of using air as the oxidizer, pure oxygen was used. This is referred to as \textit{Flameless-oxy-fuel} combustion. The extra advantages of using oxygen are the possibility of zero NO\textsubscript{X} emission, zero-CO\textsubscript{2} emission, favorable furnace atmosphere for more gas radiation, and further reduced furnace size [4]. In fact, flameless-oxy-fuel has gained a lot of attention in the last few years since it is considered as one viable potential for zero-CO\textsubscript{2} emission due to simple CO\textsubscript{2} capturing methods. To reach the same level of low oxygen concentration, the gas re-circulation rate when mixing oxygen and combustion gases is larger compared to when mixing air and combustion gases. The gas re-circulation rate, \( R \), is defined as the ratio of the mass flow rate of the recirculated combustion gases to that of the oxidizer. In addition, this high gas recirculation ratio is capable of guaranteeing the high temperature of the reactants required for the stable combustion, without the need of combination with heat recirculation. As a result, a simpler burner without a regenerator or a recuperator can be used to achieve a stable combustion of low oxygen concentration reactants when using oxygen as an oxidizer.

On the other hand, the progress in the area of HiTAC and on the designing of highly efficient regenerative air and steam pre-heaters has a significant effect on the development of the gasification process. This progress has created a gasification process with high temperature air diluted with steam called \textit{High Temperature Air/steam Gasification} (HTAG) [5]. An increase in the steam fraction in the feed gas increases the molar fraction of H\textsubscript{2} while it decreases the CO. Therefore, the heating value of the gas obtained by HTAG is higher than that obtained in conventional air blown gasification process. This medium calorific value gas is more suitable for limited pipeline distribution and for the synthesis of transport fuels such as methanol, gasoline, and commodity chemicals, due to the absence of diluents nitrogen.

Since the underlying concept of all these novel combustion and gasification technologies is the low oxygen concentration of the reactants, they are categorized under so called \textit{Oxygen-diluted Processes} (OdP). The combustion processes under this category are those characterized by reactants of low oxygen concentration and high temperature. Therefore, the term \textit{Oxygen-diluted Combustion} (OdC) is proposed be used hereinafter to refer to these combustion technologies in general. OdC includes both cases, whether the oxidizer is air or oxygen, and therefore the terms \textit{HiTAC} and \textit{flameless-oxy-fuel} combustion will indicate, respectively, to that particular case.

The flowchart in Figure 2 illustrates these classifications and the main applications related to these processes. Similar to HTAG, the application of OdC is not limited to furnaces, but can also be extended to power generation, steam boilers, incinerators, and heat engines. The homogenous charge compression ignition (HCCI) engine, for instance, is based on a concept that can be inserted in the category of Oxygen-diluted Combustion [3]. It operates at a high compression ratio until the mixture temperature is higher than that of the self-ignition temperature, so that the self-ignition occurs homogeneously in the entire chamber. The maximum temperature of the charge is kept low by means of inert dilution.

This thesis concerns only the OdC in general, with focus on the HiTAC. The dashed-line rectangle at the right bottom corner of Figure 2 includes the main scope of this thesis. Studies performed in this thesis are listed inside the open rectangles under each type of combustion, HiTAC, and flameless-oxy-fuel. Thermodynamic analysis was performed in this study for the OdC process in general. However, the study of flame characteristics and its effect on the heat transfer inside the
furnace were performed for the HiTAC cases. In addition, mathematical models were developed to simulate the regenerative heat exchangers. These models were verified by experiments performed on various commercial regenerators.

In practice, a high gas recirculation rate is achieved inside the combustion chamber by diluting the reactants with the combustion products through the use of burners with separated air and fuel nozzles, through which air and fuel are streamed at high velocities. This high velocity creates a natural entrainment effect to the air and fuel flow jets in which irrotational fluid, i.e. flue gases in the chamber, may become rotational by the diffusion of vorticity through the boundary surface of a turbulent flow, i.e. the high velocity flow of reactants [6]. In addition, the direct 'Engulfment' of the irrotational fluid by the large scale eddies is one parallel mechanism of entrainment of the flue gases.

In HiTAC, the heat recirculation is required in addition to the gas recirculation. Therefore, cyclic type regenerative burning systems are used. They operate on the principle of short-term heat storage using regenerative heat exchangers (regenerators). In the regenerators, flue gases and combustion air alternately flow through a chamber filled with a heat storage medium, e.g. ceramic, storing heat from the flue gases during the regenerative mode and releasing the heat to combustion air during combustion mode. The main functions of the regenerators are:

1- recovery of heat, 65 – 85% of the heat from the furnace waste gases;
2- pre-heating the incoming combustion air that must stream into the furnace at high-temperatures required for the HiTAC process, i.e., above the self-ignition temperature. Consequently, the

Figure 2. Flowchart showing the Oxygen-diluted Processes (OdP) and the thesis scope.

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1- recovery of heat, 65 – 85% of the heat from the furnace waste gases;
2- pre-heating the incoming combustion air that must stream into the furnace at high-temperatures required for the HiTAC process, i.e., above the self-ignition temperature. Consequently, the
thermal performance of the regenerator is an important factor to achieve this desired temperature;

3- resisting high temperatures and thermal stresses due to a high temperature gradient along the regenerator. In fact, pure Alumina is one popular material in such applications.

Regenerative burners can be classified according to the number of burners. A twin-type regenerative burner is one type (Figure 3a) in which each unit composed of a pair of burners and each burner has a regenerative heat exchanger. The burners work alternately; when one is in combustion mode the other is in regeneration mode in order to recover energy from the exhaust gases. Every certain period of time, called the switching time, they switch over. Therefore, the time duration of one cycle, cycling time, equals twice of that of the switching time. The other burning system is a so-called single-flame regenerative burner (Figure 3b) in which each unit is composed of a single burner that is comprised of at least two regenerators. In this type, switching occurs between the regenerators while the fuel continuously flows. When the single-flame burner is configured to comprise three or more regenerators, it can be sometimes that only one of them operates in combustion mode. The others that are in regeneration mode switch over to combustion mode in a serial order. In this configuration, the duration of combustion differs from the duration of regeneration in every regenerator. Depending on burner and regenerator design, the switching time is normally between 5 and 60 seconds.

The heat regenerators can also be classified in accordance to the type of heat storage medium (the packing material): either a fixed bed randomly packed with ceramic spheres or a honeycomb that is a plurality of identical flow passages. In spite of the high cost of pure Alumina, which is used as a packing material, the honeycomb can be made of composite materials in which the hot portion only is made of Alumina and the medium and cold portion is made of another cheaper material. This measure is to reduce the initial cost of the regenerator.

(a) Twin-type regenerative burner
(b) Single-flame regenerative burner

Figure 3. Schematic of twin-type and single-flame type regenerative burning systems.

The heat recirculation can also be applied to flameless-oxy-fuel systems by which thermodynamic efficiencies can be further increased.
2. **State of the art**

2.1 Thermodynamics of Oxygen-diluted Combustion

Beside the two main objectives in the development of combustion technology, which are to decrease energy consumption and to reduce pollutant emissions, there is always an interest in improving the availability of fuel energy after combustion in order to provide useful work. In fact, approximately 1/3 of the fuel energy becomes unavailable for a typical atmospheric combustion [7][8]. Most of this inherent thermodynamic irreversibility is associated with internal heat transfer between products and reactants. Daw et al. [9] demonstrated the potential for reducing thermodynamic irreversibility of combustion by considering a conceptual isobaric combustion process that utilizes controlled pre-heating to promote near equilibrium combustion. However, this was only conceptual and not very practical, but it helped illustrate the trend for less irreversible combustion.

Most thermodynamic analysis interest in Oxygen-diluted Combustion has focused on temperature issues to ensure reduced peak temperatures and temperature variations (quasi-isothermal) to achieve the above-mentioned advantages. Global thermodynamics shows that the temperature increase is reduced to a few hundreds degrees during OdC, and is lower than the inlet pre-heat temperature of the reactant mixture that is above the self-ignition temperature. Cavaliere et al. [3] shows that although in the diluted case the temperature ranges between the maximum temperature and inlet temperature, the temperatures at a diluted case will always be lower than those of an undiluted one, even if pre-heating was increased. The maximum temperature is when the reaction forms CO₂ and H₂O. Therefore, other interesting terminology to this technology was “Mild Combustion” [3] and “Quasi-isothermal Combustion” [10] because the temperature increase due to the reaction is lower or milder than that of the pre-heated reactants.

Detailed thermodynamic analysis is in the HiTAC book of Tsuji et al. [2]. They show that second law efficiency increases due to pre-heating, and is higher in the case of isothermal combustion than in the case of adiabatic combustion. However, they did not study the effect of gas re-circulation. Instead, they did analysis at different equivalence ratio, φ, from 0,1 to 1. In fact, Oxygen-diluted Combustion can be best represented by R or the reactants oxygen level, and not by φ.

However, and to the author’s best knowledge, very little attention was paid on the global thermodynamics or exergy analysis on Oxygen-diluted Combustion. The reason is probably because this technology is relatively new and most of the research was concentrated on the potential benefits of using this technology in industrial heating, in which 2nd law analysis is used only sparingly by many, and not at all by others [11].

2.2 OdC fundamentals (bench-scale studies)

The aim of the fundamental studies was to provide further insight into the thermal and chemical behaviour of the flames in the high-temperature and low-oxygen environment, which occurs when large amounts of combustion products of low oxygen concentration are mixed with combustion air, thus lowering the oxygen concentration in the reaction zone, i.e., Oxygen-diluted Combustion, for example, HiTAC.

Major issues of interest include identification of flame characteristics, quantification of the thermal field, thermal field uniformity, and emissions using various fuels. In small scale experiments, the flame thermal characteristics were obtained by measuring temperature profiles and fluctuations for cases of ambient cold air and cases of High Temperature Air Combustion (HiTAC) for comparison.
Temperatures of moderate values and smaller gradients were observed from these experiments [2], [12]. Temperature uniformity was found to be far greater with low oxygen concentration combustion air, pre-heated to ~1000°C, as compared to that obtained with ambient air at room temperature. In the HiTAC cases, temperature peaks were not present and temperature fluctuations were found to be less than 10°C compared to 200°C in the case of conventional-air combustion.

The physical characteristics of HiTAC flames, including flame shape, length, colour, and physical appearance were thoroughly investigated using bench size jet flames. Significant visual changes of the physical appearance of the flame were observed when concentration of oxygen decreased from 21% to very low levels, < 5% [13]. Flames with air having low oxygen content and high temperature had extremely low luminosity and very large volume. In the case of LPG at high temperatures, > 1000°C, and low oxygen concentration, < 5%, the flame colour was green [12] or blue [14]; the colour is bluish-green to green in the case of methane [2]. It was observed that other factors can affect the flame size as well, such as fuel jet velocity and temperature of the oxidizer. The flame becomes smaller when either of these is increased. However, the last two effects to flame size are not as high as the oxygen molar fraction [14]. In a similar study, Szewczyk [15] observed that the lift-off distance (the distance between the visible flame and the nozzle) increases with fuel jet inlet velocity increases. Moreover, higher injection velocity of the fuel lowers the emissions of NO.

However, part of the flame in which reactions are taking place is invisible and visual detection of this chemical flame is impossible. A numerical study by Yang et al. [16] showed that the chemical flame volume and chemical flame length increases with either a decrease in oxygen content or an increase in oxidizer temperature. The influence of the latter is different on the chemical flame compared to the visual flame. Furthermore, Yang found that the nozzle diameter, i.e. fuel jet velocity, has no effect on flame length. Hence, the chemical flame is different than what was visually observed during the experiments [14], [15].

Chemiluminescence’s intensity of C₂ and CH radicals from a bench scale LPG flame were measured by Hasegawa et al [12]. The C₂ and CH are indicative of the green and blue region of the HiTAC flame, respectively. The species intensities were found to increase with an increase in air pre-heat temperature above 1000°C at all wavelengths examined. The C₂/CH intensity ratio was examined and found to increase also with pre-heat temperature. Such increases are indicative of the transition from a green-blue to a green colour of flame. It was found that the C₂/CH intensity ratio is not only a function of pre-heat temperature, but also a function of the equivalence ratio. An increase in equivalence ratio (but not above 1) causes the C₂/CH intensity ratio to increase. The same investigations were carried out for methane. The results showed a significantly less increase with methane as compared to LPG (mostly propane). The C₂/CH intensity ratio, however, does not change remarkably with normal air, 21% O₂, for both fuels. For the normal temperature air flame the intensity was locally concentrated, whereas for the HiTAC case it was uniformly spread over in a combustion chamber. The emission spectra of various species in the HiTAC flame was determined at a certain location for various pre-heat temperatures > 1000°C at 4% O₂ content [17]. The results are significant because they show a significant increase of OH, CH, C₂, and H₂O emissions (e.g., the increase factor is about 2 for C₂) when the air pre-heat temperature increases slightly from 970°C to 1030°C. The extent of colour change of the flame was obtained by Gupta [17] using a specially developed computer program sensitive to colour in flame photographs. The yellow colour of the flame volume increases with an increase in O₂ concentration in the air. Moreover, the increase in flame temperature increases the yellow fraction of the flame. Experiments also showed that the flame volume associated with the green colour of the flame increases rapidly at an O₂ concentration less than 5% in combustion air.

The information regarding emissions and colours assists in providing design guidelines because the flame radiation associated with different flame colours is different. Gupta [17] also directly measured the heat flux from a bench size experimental jet flame. The results revealed a very
uniform and high heat flux in HiTAC flames due to high velocity of air in the test section, which increases the convective heat flux. In addition, the radiation is higher due to enhanced radiation heat flux from furnace walls. It was concluded that the heat flux from HiTAC flames is much higher and uniform, which can be translated into uniform heating of the material to be heated and reduced energy requirements.

Further fundamental understanding of the phenomena was brought by Mörtberg et al. [18], [19]. They studied the flow dynamics in cross-flow small jet flames. The spatial distribution of combustion intermediate species, such as OH and CH, were measured by Mörtberg et al. [18], [19], [20] using the light emission spectroscopy. The methane fuel jet in a cross-flow showed a significantly improved prolonged reaction zone and faster ignition during high temperature oxygen diluted conditions when compared to burning methane at ambient conditions. They also studied flames of LCV gas-fuel jets; the same type of measurements revealed a significant delay of the ignition of the LCV fuel jet. The mixing is hampered under high temperature air combustion. This resulted in zones of higher turbulence levels, higher axial strain rates, and higher vorticity for the low calorific fuel jets during combustion when compared to methane fuel jet. The strong heat release of methane combustion suppresses the vortex formation. This flow dynamics information was a diagnosis result of a Particle Image Velocimeter (PIV). Moreover, these experiments showed a strong dependence of the combustion air temperature and oxygen concentration on mass entrainment in the flame. The low combustion intensity caused the entrainment to increase significantly [13].

In a similar bench size jet flame, but in a co-flow jet, the entrainment under high-temperature and oxygen-deficient (HiTOD) combustion has been studied thoroughly by Yang et al [21]; however, this has been done numerically. New entrainment coefficients were determined for the oxygen diluted flames in terms of Froude number ($F_r$). The entrainment of jet flames was divided into two regions: the near field, where the entrainment coefficient is positive, and the far field, where the coefficient is negative. The effect of the heat release reduces the entrainment of the reacting jet in the near field with a factor of $(T_f/T_o)^{0.5}$. Therefore, the entrainment increases with a decrease in oxygen concentration and with a uniformity of heat release. Furthermore, it was found that it is independent of the pre-heat temperature of the oxidizer for the studied temperature range (1073-1573K). They also performed numerical and theoretical studies of OdC flames from regenerative burners in a larger scale furnace, i.e., a semi-industrial scale furnace. Among the subjects investigated were the flame shape and size, the flame entrainment, and the heat transfer uniformity [22], [23]. The CFD-based calculation and mathematical modelling used were verified using experimental data related to this thesis [24], [25]. Their work also extended to flameless-oxy-fuel [10]. When OdC was compared to conventional-air combustion, numerical results revealed reduced peak temperatures, uniform spatial temperature distribution, lower combustion intensity, larger flame volume (by 16 times), higher efficiency, and higher and more uniform thermal radiation to a heat sink (~1.7 times). It has been shown that a lower excess air ratio leads to a low peak temperature and a larger flame volume, and thus a low NO-emission. In addition, a lower excess air ratio leads to a larger flame volume and a larger flame entrainment ratio, thus lowering NOx emission further. It was proposed to use the entrainment ratio as a criterion for designing a combustion chamber [26].

As a matter of fact, when investigating new combustion phenomena, NOx emission is one of the most important issues and cannot be excluded as in most of the above fundamental studies on OdC. It is well known that nitrogen oxides form along three possible paths, namely thermal NOx, prompt NOx, and fuel NOx [27]. The first two mechanisms are effective for high temperatures. Therefore, most researchers agree that the reduced peak temperatures and uniform-moderate temperature fields in OdC flames substantially reduce the NOx formation and emissions. This was experimentally observed in all measuring occasions [2], [12], [17], [28], [29]. Gupta [17] observed that NOx emission at an air pre-heat temperature of 1150°C decreased from 2800ppm at 21% O2 to 40 ppm at
2% oxygen. Hasigawa et al. [12] presented NO\textsubscript{x} emission as a function of oxygen concentration in air and air pre-heat temperature. However, Cavaliere [3] suggested that combustion under oxygen-diluted combustion conditions is an optimum condition for NO\textsubscript{x} destruction techniques, such as reburning and selective non-catalytic reduction (SNCR). These paths or type of reactors require a very rich condition in the first combustion stage, rich presence of nitrogen atoms or cyanides and relatively narrow temperature range, around 1300K, for a relatively long residence time. In this respect, the OdC can be a suitable process.

The above small-scale experiments were of great importance in creating and enhancing knowledge about the global features of OdC combustion phenomena and identification of the main features and characteristics of the flame. These experiments provide generous information in a much shorter time with easily controlled experimental conditions. In addition, their initial and operating costs are much lower. However, these experiments are fuel jet injected into either cross-flow, co-flow, or counter flow [30] oxidizers and do not simulate the actual OdC regenerative burning system and the flames thereof.

### 2.3 OdC fundamentals (large-scale studies)

Experiments in larger scale furnaces were carried out in parallel to those in small scale test-stands in order to:

- verify the characteristics of the combustion phenomena,
- verify mathematical modelling,
- be case studies in the application of OdC technology, and, most importantly,
- quantify the main parameters related to heat transfer in furnaces and to the physical and chemical characteristics of the flame.

In the application of OdC, the importance of providing air at a very high temperature was realized and the regenerative heat exchanger gained a considerable amount of attention in the research activities mainly from burner manufacturers.

Weber et al. [31] measured local in-furnace velocity, temperature, gas composition and radiation in a semi-industrial furnace firing natural gas. The temperature and chemistry fields were uniform all over the furnace under HiTAC conditions. Spatial gas temperatures were in the range of 1350-1450°C despite the high temperature of combustion air (1300°C). The LDV measurements allowed for estimating the entrainment for the air jet and the gas jet. The air jet doubled its original mass at a distance 1 m downstream from the nozzle by entraining combustion products. The fuel gas jet with an original mass of 46 kg/hr entrained around 150 kg/hr of combustion products at a distance of 0.5 m downstream from the nozzle. This caused slow burning of methane, which was detected 2.5 m downstream the furnace front wall. The furnace resembled a large black body at a temperature of the inlet air temperature, 1300°C, because of the high heat flux (300–350 kW/m\textsuperscript{2}) measured along the whole furnace.

Among many other so-called “low-NO\textsubscript{x} burner” technologies, e.g., conventional-oxy-fuel or staged burners, Flamme et al. [32] found that at glass melting furnaces, i.e. very high temperature furnaces, burners based on HiTAC technology emit the least NO\textsubscript{x} from the exhaust gases ~500 mg/m\textsuperscript{3}. At average wall temperatures of 1500°C, the flame length from HiTAC combustion was 3.2 m compared to 2.7 m and 2.3 m for flames from a staged burner and a standard burner, respectively. This was measured according to a 2000 ppm CO concentration as an indicator for the flame boarder. A bright area of radiation from OH radicals was detected in the near burner region in all other burners. However, OH radical radiation was detected down stream of the near burner region and was distributed, indicating even and low reaction intensity. Results from radiative heat flux measurement from the flames using the narrow angle radiometer indicate that the heat flux from the
HiTAC flame was slightly less than that from other flames, probably due to the reduced flame temperature. The measured average flame thermal radiation was highest from the standard flame. However, the total radiation fluxes incident, measured using the ellipsoidal radiometer, indicate that the heat flux from the furnace using HiTAC burner was in the same range as those using other burning techniques.

In fact, the regenerative burners were NOT used in the above two works [31], [32] and were simulated with a burner operated under steady state conditions to avoid the dynamic operation of a real regenerative burner in order to make these measurements possible. In spite of this fact, the accuracy of velocity measurement was no better than 20-30%.

Awosope and Lockwood [33] performed modelling-based theoretical study on three different semi-industrial furnaces. They found out that the percentage of the total heat release radiated by HiTAC-flame for all cases exceeded the range of 10-15% quoted for radiative heat transfer in the conventional flames of natural gas, i.e., the same fuel used in their study. The principal consequence of the high radiant heat loss is that the energy release is significantly moderated, thereby resulting in minimized temperature rise and a relatively uniform temperature field. The local temperature and species concentration profiles were measured and showed agreement with the results.

The above furnaces were operating under HiTAC condition, resembling a well-stirred reactor [12], [31], [33] due to the uniformities of temperature, heat flux, and chemistry fields all over the furnace. In fact, some argue that OdC is diffusion combustion, although it is volumetric combustion rather than flame front combustion [16].

Suzukawa et al. [34] showed that it is possible to burn low calorific value fuels using HiTAC technology in a semi-industrial test furnace. Fuel gas, steel works by-product gases of 11.5 MJ/m³, was fired in a test furnace at 0.93MW using two pairs of twin-type regenerative burners. NOx emissions remain below 50ppm at furnace wall average temperature of 1350°C. They found that the amount of energy savings is not only from the energy gained by pre-heating air using heat regenerators but also from the increased heat potential of the furnace gases due to this pre-heat. Heat transfer CFD-based calculations for a slab reheating furnace revealed a 40% fuel savings when retrofitting a conventional reheating furnace equipped with a cold air combustion system with a new HiTAC regenerative burning system. The fuel savings was 10–15% for retrofitting recuperators. Zhang et al. [35] developed a mathematical model for steel slab reheating furnaces equipped with HiTAC regenerative burners. An agreement between predicted gas temperatures and measurements was found.

Case studies showed that HiTAC technology was applicable and beneficial in most industrial furnaces. The HiTAC technology was applied to one of the most consuming energy industrial processes, steam reforming [36]. In this pilot project, innovation of design concepts was made for both the process configuration and the steam reformer thereof. The innovation included downsizing the waste heat recovery by using a relatively small size single-flame HiTAC regenerative combustion system [37] and downsizing the reformer itself. These innovations would realize downsizing equipment, reducing energy consumption, and minimizing environmental pollutants. The newly invented combustion system based on HiTAC, which was responsible for the downsizing of the reformer, provided uniform heating along and across the tube and reduced NOx emission to 20 ppm (corrected 6% O2). The use of the ionization technique to detect invisible OdC flames was behind this development [38]. The ionization detection also contributed in enhancing the heating effect by reducing temperature deviation across the tubes from 50°C to 15°C and reducing temperature deviation along the tubes surfaces to 100°C, with 20 ppm of NOx emission. Moreover, other important benefits were also realized when using this advanced reforming process based on HiTAC technology, e.g., prolongation of the tubes life and product efficiency.

In a three year period in Japan, 1998, 1999, and 2000, 167 plants converted to HiTAC and resulted in saving 156 M liter oil-equivalents per year [39]. Case studies of early application of the HiTAC
in different types of furnaces in Japan created considerable interest among industries in Europe [40], [41]. Some examples include, with mention to the energy savings without other benefits:

- Billet reheating walking beam furnace using recuperators and having a 100t/hr throughput capacity: the specific energy consumption was reduced from 1.1 to 0.88 GJ/ton when retrofitting the burners with HiTAC-based twin-type regenerative burners.

- Aluminum melting furnace having a 20 ton/hr throughput capacity using heavy oil: the energy savings was about 47% when retrofitted with a half-capacity HiTAC-based burning system firing natural gas. CO2 emission reduction was consequently higher than that.

- Batch type gas carburizing furnace using two radiant tubes with a 600 kg/charge throughput capacity: energy savings was around 23% as a result of retrofitting with HiTAC-based regenerative burners.

In Europe a few examples of HiTAC firing systems were published in annealing and pickling line furnaces [42] and in strip line furnaces using radiant tubes [43]. A distinction between re-circulating and non-re-circulating tubes was made. Re-circulation type radiant tubes can be installed whenever non-recirculating tubes are in operation for enhanced gas-recirculation. Additional expenses for a HiTAC-based efficient radiant tube system can be recovered in a few years. Counting other advantages, especially increased production, less downtime, temperature uniformity, and a longer tube lifespan can reduce the payback period for these systems. However, no quantification information is available regarding heat flux intensity or uniformity from these radiant tubes.

Despite the increased number of flameless-oxy-fuel applications in the industry in Sweden [44] compared to HiTAC technology, only a few investigations were carried out in this field in order to compare flameless-oxy-fuel with HiTAC and other technologies [10] [44]. This subject requires further and thorough fundamental investigation.

The above large-scale experiments were carried out by many researchers [31] [32] in order to investigate the thermal and chemical characteristics of Oxygen-diluted Combustion (OdC). However, the following was observed:

- Regenerative burners impose transient conditions of furnace operation due to their cyclic firing configuration, making simple measurement difficult. Therefore, they were not used and instead were simulated with burners operated under steady state conditions. In spite of this fact, the accuracy of velocity measurement, as an example, was no better than 20-30% [31].

- Available quantitative information about radiative heat flux measurements is still sparse.

- In spite of the above research activities, the convective mode was almost always regarded as insignificant and rarely investigated in furnaces operating at high temperatures.

- Available quantitative information regarding thermal performances and efficiencies of the furnace are originated from burner manufacturers’ technical reports [28], [34], [37], [40].

- These were experimental investigations that ignored the dynamic operation effect of the measurements.

- In a real industrial furnace, the efficiency was calculated by means of the specific fuel consumption based on the long-term fuel and production metering system. This type of measurement normally ignores the number of operational interruptions due to maintenance and production requirements.
2.4 Heat regenerators in HiTAC burning systems

Heat regenerators do not only reduce the fuel consumption in a HiTAC burning system but also provide the necessary high temperature of air required for combustion. Consequently, the thermal performance of the regenerator, which is probably the most important element of the whole burning system, is an important factor to achieve the desired temperature of the combustion air. The heat exchange efficiency depends mainly on the regenerator structure, heat transfer properties of the regenerator material, switching time, flue-gas suction rate, and others.

The classical books by Hausen [46], Schmidt and Willmott [47], and Kays and London [48] provide excellent background on analytical methods and mathematical models for calculating temperature profiles, effectiveness and other thermal performance parameters of regenerators having different heat transfer geometries, and flow patterns. Simple design techniques such as $\Lambda$–$\Pi$ and $\varepsilon$-NTU were developed and presented in these books.

The first approximate solution to determine the spatial temperature distribution in solid material at a certain time and at a certain cross section in a heat regenerator was presented by Heiligenstaedt and Rummel [46]. They assumed a time independent of fluid flow temperature, an assumption that does not hold well in practice. In order to get away from this assumption Rummel introduced some constants to the heat transfer coefficient that were determined by experiment. There was another approximate solution developed by Schack, in which he formed empirical expression that also contained constants that were experimentally determined. These methods were restricted to a brick or plane wall cross section. In addition, Hausen [46] developed a theory that was free from the above assumptions. He used the zero Eigen value function, which corresponded to the fundamental temperature oscillation. However, this theory was only valid when thermal capacities and switching times were equal for both gases. His theory was extended to cylindrical and spherical shapes as well.

Efficiency and complete temperature distribution between the entrance and the exit of a heat regenerator can also by determine using the graphical method, $\Lambda$–$\Pi$ [46], [47], [49], [50], which is a consequence of massive numerical calculations used to solve integral equations that are based on the zero eigen function for the limiting case of identical thermal capacities and switching times for both gases. The $\varepsilon$-NTU method [48] is another simple design tool, but it does not predict the temperature distribution in solid materials or gases. Trade-off factors to weigh the relative costs of pressure drop, weight, heat transfer performance, and leakage in the case of regenerative heat exchangers were developed. In fact, infinite thermal conductivity, $k = \infty$, perpendicular to flow direction of solid material was assumed in both methods. Consequently, a severe disagreement was observed when comparing solid surface temperature between an infinite $k$ solution with a finite one, even at $\text{Bi} = 0.1$ [47], especially at the entrance regions. Moreover, all described methods so far assume that the thermal properties of solid materials and gases are constants, although the heat transfer coefficient is strongly dependent on temperature. In addition, they neglect the gas axial thermal conductivity, parallel to the flow, and the energy ratio between the gases entrained in the storage material and the material itself. The later effect is not small for compact regenerators operating at short switching times.

An approximate close analytical solution for regenerative heat exchangers with matrices in the form of a thin-walled monolith was developed by Klein et al. [51]. It considered the axial heat conductivity of the storage material. End temperatures and effectiveness of this solution agreed approximately with numerical simulation models for a fast switching limiting case. However, less agreement was observed when temperature profiles along the regenerator were compared to numerical simulations.

Alternatively, two-dimensional numerical simulations based on finite element methods can take into account the thermal conductivity parallel and perpendicular to the flow direction of gas and the
effects of gas heat capacity entrained in the regenerator. Moreover, the temperature dependent thermal properties can be accounted for, and hence simulate composite material regenerators without difficulties. They provide a more precise calculation of temperature distribution along the regenerator. In contrast, one-dimensional numerical simulation, as described by Tada et al. [52], is not capable of considering all the above effects. One dimensional model are still useful for the prediction of temperature profiles in fixed bed regenerators [53], [54], although none have been applied on regenerators used in HiTAC burners or applications.

Sudoh et al. [40] and Mochida et al. [37] are some of few who investigated, theoretically and experimentally, regenerators that are especially used in HiTAC applications. They studied the effect of switching frequency in regenerative combustion on the heat transfer rate and temperature efficiency. Suzukawa et al. [34] measured the regenerator efficiency for various flue-gas suction rates (FSR). FSR is defined as the amount of flue gases pulled from the furnace to preheat a unit of combustion air. However, the effect of the dynamic behaviour of the heat regenerators operation on the measurement data was ignored throughout the experimental work. Methods for considering this effect on measurements in unsteady state conditions have been treated by Khalil E. [55].

For Oxygen-diluted Combustion based on oxy-fuel application, i.e., flameless-oxy-fuel, although the use of regenerators would reduce flue gas energy losses, thereby reducing energy and oxygen consumption, it is probably still uncommon for the following reasons: (i) the absence of nitrogen reduces exhaust gas energy loss, (ii) a simpler and cheaper burner without a regenerator or a recuperator can be used to achieve a stable combustion due to the high gas re-circulation ratio, and, most importantly (iii) the maximum heat recovery rate from the oxy-fuel flue gas is limited to about 40% [56] because the ratio of flow-stream capacity of oxygen to that of the oxy-fuel flue gas is much smaller than that in an air combustion process.

To maximize the energy recovery from flue gases produced by oxy-fuel combustion, a ‘thermochemical regenerative heat recovery’ by which a significant fraction of the thermal energy stored in a regenerator is recovered was recently patented [57]. In this process the high concentrations of H₂O and CO₂ in the flue gas of oxy-fuel combustion provide a unique advantage. Natural gas is mixed with recycled flue gas and reformed in hot regenerative beds to H₂ and CO through endothermic reforming reactions. Furthermore, although the question seems to be economical in the first place, the effects of heat recirculation in an-industrial scale furnace based on flameless oxy-fuel have to be investigated.

2.5 Development of OdC technology

Well-known achievements of the NEDO’s (The New Energy and Industrial Technology Development Organization, Japan) research programmes on Oxygen-diluted Combustion (OdC) performed under the leadership of Tanaka [28], resulted in the development of excess enthalpy combustion to a combined heat and gas recirculation combustion that was coined as “Ultra-high pre-heated air combustion”. Significant advantages were realized of such combustion, e.g., excellent combustion stability in a very wide range of air/fuel ratios (0.5<λ<5), low noise, low NOx emission, uniformity of temperature and heat flux, energy savings by waste heat recovery, and increased heat transfer. Tanaka also described the burning system that was innovated, the twin-type regenerative system, and the techniques for how realizing HiTAC in an industrial steel heating furnace and in a radiant tube. Parallel research activities that took place in Germany resulted in adapting the same combustion concept as an effective technique for reducing NOx emissions. Wünning et al. [29] found out that under certain conditions, such as high recirculation rates, the combustion took place without any visible or audible flame, and hence the term “flameless combustion” was coined. Under flameless conditions, no high temperature peaks or high gradients of temperatures and species exist, thus suppressing NO formation even at very high air pre-heat temperature. Wünning summarises
several NOx reduction techniques and showed that the NOx emission from burners operated in flameless oxidation mode can be reduced by one or two orders of magnitudes from that produced by a typical low-NOx air staged, high velocity burner. Industrial furnaces and radiant tubes utilizing such burners have proven to be reliable for very enhanced and uniform product quality as well. These innovations created a great deal of interest and knowledge on such novel combustion technology.

In Japan, the technology was qualified as one reliable technology in industrial furnaces for dramatically reducing energy consumption and consequently GHG emissions (~30%). Therefore, its applications were disseminated and subsidized by the Japanese government in 1998 and 1999 [58]. Industries, research institutes, and the energy agency in Sweden (STEM) showed a great deal of interests on HiTAC as well. Several research activities were lunched and carried out to investigate this technology fundamentally in a bench scale experiment [59] and to explore its benefits for the Swedish industry [60], [61]. In fact, the Swedish industry took one step forward by utilizing flameless-oxy-fuel technology instead of conventional-flameless, or HiTAC. In the Swedish steel industry, a total power of 220 MW and 100% oxy-fuel combustion systems were in daily operation in 56 heating furnaces in the year 2003 [44]. In fact, due to the implications of the Kyoto protocol on combustion, CO2 capture, and storage, flameless-oxy-fuel has become one important option for GHG mitigation. Therefore, oxy-fuel with flue gas recirculation and flameless-oxy-fuel gained great importance and interest related to combustion research and applications [62]. The state-of-the-art air separation technology requires about 220 kWh of electric energy to produce a metric ton of gaseous oxygen at 1 atm and at 95% O2 purity [56]. The overall energy efficiency of the oxy-fuel combustion process, including the energy required to produce oxygen by conventional air separation processes, often becomes higher than that of an air fired combustion system even if waste heat recovery techniques were used. In addition to the qualitative and quantitative benefits of flameless-oxy-fuel compared to HiTAC, simple and less expensive CO2 capturing methods can be applied to oxy-fuel combustion gases.

2.6 OdC in process industry and work conversion

In the application of power generation and steam boilers, the concept for a new HiTAC-based boiler was designed in which most of the convective parts are replaced by radiative parts, hence downsizing the boiler [28]. To the best knowledge of the author, the utilization of HiTAC technology in boilers does not exist yet. However, the use of flameless-oxy-fuel in boilers has already started, which indicates the big potential of such technology. Alstom developed oxygen-fired CFB uses pure oxygen tempered with re-circulated flue gas to combust the fuel [63]. Pilot tests have confirmed the technical feasibility of an oxygen-fired CFB system. Without the requirement of CO2 sequestration, oxygen-fired CFB can be economically feasible where the byproducts, CO2 and N2, are sold for enhanced oil recovery.

In internal combustion engines, the homogenous charge compression ignition (HCCI) engine is based on a concept that can be inserted in the category of Oxygen-diluted Combustion, since the maximum temperature of the charge is kept low by means of inert dilution [3] or by exhaust gas-recirculation [64]. This allows the engine to operate at a high compression ratio until the mixture temperature is higher than that of the self-ignition temperature, so that the self-ignition occurs homogenously in the entire chamber. The high compression ratio means higher efficiency. However, the burning velocity is decreased due to gas recirculation in a constant volume combustion chamber. Therefore, an adjustment to the fuel by adding hydrogen or a reform gas will raise the burning velocity back to an undiluted level [64]. Moreover, the features of Oxygen-diluted Combustion technology include a mild increase of temperature, low-noise, etc., and are favorable in the application of gas turbines, although very few GT systems have been built with the purpose of satisfying the OdC condition.
3. Objectives

The objectives of this thesis are:

1- Performing thermodynamic analysis of Oxygen-diluted Combustion technology for systems, utilizing either air or oxygen as an oxidizer to explore the potentials for reduced thermodynamic irreversibility and to examine the effect of oxygen-diluted conditions in the temperature increase due to combustion.

2- Quantifying the enhancement in efficiency resulted by HiTAC in order to explore potentials for reduced energy consumption in industrial furnaces by:
   – performing energy balance measurements on direct- and indirect-heating industrial furnaces and various heat regenerators, and
   – developing and experimentally verifying mathematical models for the two main types of heat regenerators used in HiTAC burning systems, i.e., honeycomb and fixed bed regenerators.

3- Quantifying heat transfer enhancement due to HiTAC inside a direct and indirect-heating industrial furnace by carrying out local measurements in a semi-industrial test furnace equipped with industrial HiTAC regenerative burner systems in order to establish combustion and heat transfer conditions prevailing in an industrial furnace. Among the local measurements are temperature, temperature fluctuation, flue gas composition, and radiative and convective heat fluxes. This is because a complete understanding of the heat transfer in industrial furnaces has always been required in the design and the optimization of heating processes.
4. Experimental facilities and measurements

A semi-industrial furnace equipped with state-of-the art burners and measuring equipments was mainly used to carry out the experiments. Tests were also carried out on a fixed bed regenerative burning system that was mounted on one steel reheating furnace at SSAB TUNNPLÅT AB. This chapter describes these facilities and highlights the main features that are important for every specific investigation.

4.1 Experimental set-up of a semi-industrial test furnace.

The semi-industrial test furnace was built in 1999 [65] at the Royal Institute of Technology (KTH), division of Energy and Furnace Technology (EFT). The outer dimensions of the furnace body were 3.5 x 2.2 x 2.2 m. The test stand was built for a maximum firing rate of 500 kW (Figure 4). The furnace body was isolated with a 0.3 m thick layer of ceramic fiber material. There were also a number of openings in the furnace body for observation and measurement inside the combustion chamber (Figure 5). Heat was taken away from the furnace with four fixed horizontal air-cooling-tubes made of a special temperature resistant Kanthal® alloy. They provided a large range of load to be removed from the furnace. Furthermore, the test furnace was equipped with two different flue gas channels. One was for hot flue gases equipped with a water-cooled heat exchanger, and the other was for cold flue gases from the heat regenerators. The flue gas channel for hot gases allowed the furnace to be run in conventional mode or in modes in which part of the exhaust gases passes through the regenerator.

Figure 4. Photos of the semi-industrial test furnace at KTH. The burners are shown in the photos on the left and the traversing system with the water cooled probe is shown in photo of the roof of the furnace.
The furnace was equipped with two different types of regenerative systems: a ‘single-flame’ system and two ‘twin-type’ systems, each consisting of a pair of burners (four burners in total). Both are commercially used and based on the honeycomb compact heat regenerators. The positions of the different burners in the experimental facility can be seen in Figure 4 and Figure 5.

In the single-flame regenerative burning system (Figure 3b), there were six regenerators in which three were in combustion mode when the other three were in regeneration mode. The switching time of this system is fixed to ten seconds, and FSR cannot be more than 80% due to design limitations. The firing capacity of this burning system is 200 kW.
The twin-type regenerative burners have a capacity of 100kW/pair. The dimensions of the honeycomb heat storing material are 150W X 150H X 300L. The flue gas suction rate (FSR) is normally 100% and the switching time can be set to a value between 15 and 60. The honeycomb matrix size is attributed to 100 cells/ in² (Figure 6). The specific heat transfer area, \( a_s \), in this case was 4200 m²/m³, and the specific weight was 0.084 kg/kW, i.e., five times less when compared to a typical fixed bed packed with balls regenerator.

In addition to the above burners used for direct heating combustion systems, a W-shape Radiant-tube (RT) was mounted vertically in the furnace (Figure 8f) to carry out experiments related to HiTAC in an indirect-heating system. The outer diameter of the tube was 0.195 m and its maximum firing rate was 200 kW. The RT was fired using two different burning systems, either a recuperative burner or a pair of twin-type regenerative HiTAC burners. The nominal firing capacity of the regenerative system was 160 kW/pair. It had a 300 mm long cylindrical regenerator (Figure 7), having the same cell structure as in the previous honeycomb. The operation of this system required some of the combustion air to pass by the regenerator, making the FSR value between 130–140%. These twin burners were placed at both ends of a W-shape radiant tube. Details about the radiant-tube and the twin burners are described in detail in Supplement V.

4.2 Firing configurations

Using the above state-of-the-art test facilities, seven different operating and firing configurations were possible for operation. The first configuration was conventional combustion (Figure 8a) in which ambient air at ambient temperature is burned with the gas fuel. This configuration was tested in order to compare it with other HiTAC regenerative systems. The second configuration was the HiTAC single-flame regenerative burner (Figure 8b). In fact, this burner was the same burner that was used for conventional combustion, the 1st configuration.

The twin-type regenerative burning system can be operated in three different configurations of operation, namely counter, parallel, and stagger, depending upon the selection of the individual burners for each pair. In counter configuration (Figure 8c), the pair of burners, A and B, comprise one unit, and burners C and D comprise another. The parallel and stagger configurations are self explained when looking at Figure 8d and Figure 8e, respectively. The last two configurations are related to the RT. A recuperative radiant type burning system (Figure 8f) with a nominal firing capacity of 175 kW, was used to represent a conventional system for comparison with the HiTAC regenerative system (Figure 8g) with a nominal firing capacity of 160 kW. All of the above burners were fired using LPG gas.

4.3 Fixed-bed twin-type regenerative burner

In addition to the semi-industrial test furnace and the burners thereof, experiments were carried out using another facility, i.e., a twin-type regenerative burning system having a fixed-bed regenerator randomly packed with spheres, 14 mm in diameter. This system was installed in a steel slab pre-heating furnace in an industry in Sweden [60]. The system capacity was 2 MW/pair and, contrary to all other burners, it burns heavy oil number five (87% carbon and 12.3% hydrogen). The standard switching time was 67 seconds. It can be set to other values but not more than 77 seconds. The bed size is 700 W X 1250 L X 510 H mm. The specific heat transfer area was 265 m²/m³, and the specific weight was 0.5 kg/kW. Figure 9 shows the system and the outside dimensions of the balls room.
4.4 Regenerative heat exchangers

Experiments on the above fixed-bed regenerative system concerned only the thermal performance of the heat regenerator and performed to verify a mathematical model that was developed in this work. With other burning systems, a total of four different regenerative heat exchangers were examined. Specifications and important properties of these regenerators are tabulated in Table 1.
Table 1. Specifications for regenerators used in experiments.

<table>
<thead>
<tr>
<th>Regenerator no.</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burning system</td>
<td>twin</td>
<td>twin</td>
<td>twin</td>
<td>single-flame</td>
</tr>
<tr>
<td>Regenerator</td>
<td>fixed bed</td>
<td>honeycomb</td>
<td>honeycomb</td>
<td>honeycomb</td>
</tr>
<tr>
<td>Packing materials</td>
<td>alumina</td>
<td>alumina/cordierite</td>
<td>alumina/cordierite</td>
<td>alumina</td>
</tr>
<tr>
<td>Application</td>
<td>reheating furnace</td>
<td>reheating furnace</td>
<td>W-shape radiant tube</td>
<td>reheating furnace</td>
</tr>
<tr>
<td>Burner nominal capacity</td>
<td>2 MW</td>
<td>100 kW</td>
<td>160 kW</td>
<td>200 kW</td>
</tr>
<tr>
<td>Matrix structure</td>
<td>$\phi = 14 \text{ mm}$ spheres</td>
<td>100 cell/in$^2$</td>
<td>100 cell/in$^2$</td>
<td>100 cell/in$^2$</td>
</tr>
<tr>
<td>Regenerator dimensions (mm)</td>
<td>700W</td>
<td>150W</td>
<td>145$\phi_o$</td>
<td>86 $\phi_o$</td>
</tr>
<tr>
<td></td>
<td>1250H</td>
<td>150H</td>
<td>80 $\phi_i$</td>
<td>27 $\phi_i$</td>
</tr>
<tr>
<td></td>
<td>510L</td>
<td>300L</td>
<td>300L</td>
<td>550L</td>
</tr>
<tr>
<td>$a_s$ (m$^2$/m$^3$)</td>
<td>265</td>
<td>4200</td>
<td>4200</td>
<td>4200</td>
</tr>
</tbody>
</table>

4.5 Measuring and diagnostic Equipments

For the semi-industrial furnace, temperatures and flow rates of all streams entering and leaving the furnace were measured to perform the furnace energy balance. The same applies to the heat regenerators by adding differential pressure measurements across the regenerator.

For the furnace local measurements, the measurements were intrusive and performed using several meters and equipments. The meters were held inside water-cooled probes. The probes were mounted at the top of the furnace and moved by a computer-controlled traversing system. They were inserted at various points inside the furnace through the 13 openings at the furnace roof (Figure 5). In each opening the measurements were performed at every 100 mm from the ceiling down to the burner centerline. At locations close to the reaction zones, distances between measuring points were even less (down to 25mm), especially when measuring flue gas composition.

In fact, the dynamic operation imposed by the switching devices of the regenerators creates one major measuring difficulty due to the inertia of the measuring devices. Therefore, several measures were taken during and after measurement to overcome this problem. This also applied during and after the measurements of the heat regenerators.

These meters were:

1- Suction pyrometer to measure the local gas temperature. Between the S-type thermocouple and a double layer ceramic shield, the gases passes at high velocity to increase heat transfer by convection. The ceramic shield tremendously reduces thermal radiation exchange between the hot environment in the furnace and the thermocouple.

2- Thin-wire thermocouple to measure local fluctuations in temperature. The junction is only 80 $\mu$m in diameter and thereby has a very short response time, time constant = 25 ms [66].

3- Ellipsoidal radiometer, to measure the irradiation [67], i.e., radiation heat flux incident per unit area falling on a surface, $G_c$. The radiation pyrometer is constructed so that the incoming radiation enters a small orifice at the tip of the probe and is focused by a mirror onto a thermopile, which gives a potential difference as a function of the radiative energy received (Figure 10). A small flow of nitrogen inside the cavity of the radiation pyrometer prevents the entry of small particles and combustion gases that would generate condensation and deposits of
liquid and solid particles on the mirror on the cavity walls. The mirrors on the cavity wall are coated with a highly reflective material such as polished copper or gold. This meter was first proposed by Nils-Erik Gunners in 1967 [68].

The mirrors on the cavity wall are coated with a highly reflective material such as polished copper or gold. This meter was first proposed by Nils-Erik Gunners in 1967 [68].

Figure 10. Ellipsoidal radiometer.  
Figure 11. Total heat flux meter (plug-type).

4- The plug type total heat flux meter to measure the rate of heat flow per unit area on the meter surface [67]. The construction of this meter is illustrated in Figure 11. It consists of a cylindrical metal plug that is insulated by a thin air gap in the lateral sides and cooled at only the rear surface. During measurements, the front surface of the plug is normally facing the hot environment, i.e., a combination of the combustion flue gases, furnace walls, and/or flame. The total heat flux, \( q_T \), received by the surface by convection and radiation, \( q_c \) and \( q_r \), respectively, is used to heat the plug by conduction, \( q_{\text{cond}} \), which can be measured by the temperature gradient along the plug. The temperature gradient can be measured by a thermopile or two thermocouples, one is located close to the rear, cold, surface of the plug and the other is located close to the front, hot, surface of the plug. The meter assembly is mounted at the tip of a water cooled probe. In fact, this total heat flux meter serves to complement the data obtained by radiative heat flux and gas temperature, \( T_g \), measurements in order to obtain the convective heat transfer coefficient, \( h_c \).

The above two heat flux meters can be calibrated in a black body furnace in accordance to several international standards. e.g., Nordtest Method NT Fire 050.

5- Gas sampling probe and micro-gas chromatograph: the probe quenches the sample down to below 140 °C in a very short time. Several measures were taken regarding sample conditioning to cope with the problem of dynamic operation. Components such as coolers and filters were selected to have as small a size as possible to reduce sample travelling time. These components were also selected from a type that ensures plug flow of the sample, and to avoid as much as possible a variation of sample composition during traveling from the probe tip to the analyzers.

The samples from this probe were then analysed using a micro gas chromatograph to measure the molar fraction of \( H_2, O_2, N_2, CH_4, CO, CO_2, \) and hydrocarbons up to \( C_4 \). The micro GC [69] consists of up to four independent modules, i.e., independent GCs that the components thereof are micro machined and small in size, providing speed in analysis. To further reduce analysis time, each GC module could be configured with a pre-column and a back-flush. The use of a GC would not be possible in such experiments without this high analysis speed, which was around 40 seconds. The micro-GC was also used in measuring the gas fuel compositions.

\( NO_x \) was measured using a chemi-luminescent analyser, by measuring the excitation of electronic energy states by the chemical reaction of nitrogen monoxide and ozone [70].
Radiant tube temperature profiles measuring points

In the case of an indirect heating system, the aimed quantification of the heat flux intensity and uniformity from a radiant tube was measured by the temperature profile of the tube. Therefore, Seventy-four K-type thermocouples were placed at 41 locations along the length of the tube (Figure 12). At least one thermocouple was mounted at each location on the right side of the tube. Points marked with 2s indicate where two thermocouples were mounted, one on the left and one on the right side of the tube. The locations that were close to the burners are marked as 4s to indicate that there were four thermocouples mounted on the left, right, top, and bottom sides. At the elbows marked with 3s, there were three thermocouples mounted on the right, left, and on the surface with the smaller radius. It was not possible to mount a fourth thermocouple at the elbows because of interference with the tube supports. The tube temperature reference point (TRP), was located at a distance at about 1 m from the beginning of the working part of the tube (Figure 12).

Figure 12. Right side of RT and locations of temperature measurement points on the tube wall.
5. Methodology of experimental work

This chapter focuses only on the methodologies, procedures, and conditions of the experiments; although the study has been theoretical as well, such as 2nd-law thermodynamic analysis and developing mathematical models related to heat regenerators. Methods regarding these theoretical investigations are explained in the next chapter.

These experiments were carried out in order to achieve the second and third goals of this work. Various types of regenerative burners were used at various burner configurations, including direct and indirect firing systems, i.e., radiant tube. Therefore, a set of intense measurements and analysis were carried out:

- conducting energy balance and heat performance measurements of the semi-industrial test furnace, the radiant tube system, and the various types of heat regenerators;
- conducting local measurement of heat-fluxes, temperature, temperature fluctuation, and flue gas composition inside a semi-industrial test furnace;
- analyze and characterize physical properties of HiTAC flame in the furnace using the above measurements and the global thermodynamic analysis;
- examine the effect of this flame on the heat transfer inside the furnace;
- quantify the radiative and the convective heat transfer from the flame; and
- in the case of an indirect heating system, measure the temperature profile of a radiant tube and quantify the heat flux intensity and uniformity from the tube.

5.1 Experimental operating conditions

Open-flame firing (direct heating systems)

The measurements were performed with one conventional air-combustion and four different regenerative HiTAC configurations according to the first five configurations of Figure 8. The four HiTAC cases were single-flame, twin-flame counter, twin-flame parallel, and twin-flame stagger. Table 2 lists the operating conditions of each configuration. The furnace temperature is an average wall temperature measured using six thermocouples, three along one side and three along the ceiling of the furnace, and was kept at 1100°C for all tests. In all cases, the firing rate and the furnace temperature, and cooling-tubes surface area were all kept constant. The furnace temperature is controlled by varying the cooling air flow rate.

Table 2. Operational conditions in the five studied cases.

<table>
<thead>
<tr>
<th>Configuration</th>
<th>$V_E$ (Nm$^3$/h)</th>
<th>$V_d$ (Nm$^3$/h)</th>
<th>$T_a$ (°C)</th>
<th>$t_s$ (sec)</th>
<th>Type of local measurements</th>
</tr>
</thead>
<tbody>
<tr>
<td>1- Conventional</td>
<td>7.7</td>
<td>200</td>
<td>25</td>
<td>-</td>
<td>$T_g$, $Gr$, $GC$, $q_T$</td>
</tr>
<tr>
<td>2- single-flame HiTAC</td>
<td>7.7</td>
<td>210</td>
<td>~940</td>
<td>10</td>
<td>$T_g$, $Gr$, $GC$, $q_T$</td>
</tr>
<tr>
<td>3- twin-flame HiTAC counter</td>
<td>7.7</td>
<td>210</td>
<td>~1040</td>
<td>30</td>
<td>$T_g$, $Gr$, $GC$, $T_{fluc}$</td>
</tr>
<tr>
<td>4- twin-flame HiTAC parallel</td>
<td>7.7</td>
<td>210</td>
<td>~1040</td>
<td>30</td>
<td>$T_g$, $Gr$, $T_{fluc}$</td>
</tr>
<tr>
<td>5- twin-flame HiTAC stagger</td>
<td>7.7</td>
<td>210</td>
<td>~1040</td>
<td>30</td>
<td>$T_g$, $Gr$, $T_{fluc}$</td>
</tr>
</tbody>
</table>
Local measurements for each configuration, according to the last column, were carried out. These measurements were total heat flux, $q_T$, radiative heat flux, $G_r$, flue gas composition, GC, temperature, $T_g$, and temperature fluctuation, $T_{fluc}$.

Radiant tube (indirect firing systems)

The last two configurations in Figure 8, the conventional recuperative system (CRS) and the HiTAC regenerative system (RS) were used in these tests. The investigations were made at eight test operating conditions, four for each configuration for comparison (Table 3). The operation was at a fixed excess oxygen content (3% O₂) for all tests, at four different levels of tube reference point temperature, TRP, (840°C, 880°C, 950°C, and 1000°C) and at three levels of firing capacity, $\dot{Q}$, (78, 129 and 155 kW).

### Table 3. Operating conditions of tests related to the radiant tube.

<table>
<thead>
<tr>
<th>no.</th>
<th>Test no.</th>
<th>Configuration</th>
<th>Fuel flow rate (Nm³/h)</th>
<th>Firing power (kW)</th>
<th>TRP temperature (°C)</th>
<th>Switching time (seconds)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>I</td>
<td>CRS1 conventional recuperative</td>
<td>3</td>
<td>78</td>
<td>840</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>I</td>
<td>RS1 HiTAC-based regenerative</td>
<td>3</td>
<td>78</td>
<td>840</td>
<td>30</td>
</tr>
<tr>
<td>II</td>
<td>II</td>
<td>CRS2 conventional recuperative</td>
<td>5</td>
<td>129</td>
<td>880</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>II</td>
<td>RS2 HiTAC-based regenerative</td>
<td>5</td>
<td>129</td>
<td>880</td>
<td>30</td>
</tr>
<tr>
<td>III</td>
<td>III</td>
<td>CRS3 conventional recuperative</td>
<td>5</td>
<td>129</td>
<td>950</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>III</td>
<td>RS3 HiTAC-based regenerative</td>
<td>5</td>
<td>129</td>
<td>950</td>
<td>30</td>
</tr>
<tr>
<td>IV</td>
<td>IV</td>
<td>CRS4 conventional recuperative</td>
<td>6</td>
<td>155</td>
<td>1000</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>IV</td>
<td>RS4 HiTAC-based regenerative</td>
<td>6</td>
<td>155</td>
<td>1000</td>
<td>30</td>
</tr>
</tbody>
</table>

Regenerative heat exchangers

For the four different regenerative heat exchangers, measurements of temperatures in the cold and hot sides of the regenerators, flow rates of combustion gases and combustion air through regenerators, and composition of combustion gases were carried out at the following various operating conditions summarized in Table 4:

- different flue gas suction rate, i.e., different capacity rate ratio $C_a/C_g$. $C_a/C_g$ is typically ~0.9 when all flue gases pass through the regenerator for LPG firing systems;
- different types of heat storage materials, honeycomb, and fixed bed;
- different switching times, i.e., different $C_r/C_a$. The matrix capacity rate, $C_r$, is dependent on cycling time and expressed as $C_r = (m_i) (c_i) (cycles/hr) (1/3600)$, where $m_i$ is the mass of both matrices.

### Table 4. Specs and test conditions for burners and regenerators used in experiments.

<table>
<thead>
<tr>
<th>Regenerator</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burning system</td>
<td>twin</td>
<td>twin</td>
<td>twin</td>
<td>single-flame</td>
</tr>
<tr>
<td>Regenerator</td>
<td>fixed bed</td>
<td>honeycomb</td>
<td>honeycomb</td>
<td>honeycomb</td>
</tr>
<tr>
<td>Packing materials</td>
<td>alumina</td>
<td>alumina / cordierite</td>
<td>alumina / cordierite</td>
<td>alumina</td>
</tr>
<tr>
<td>Firing capacity (kW)</td>
<td>2000</td>
<td>200, 174 &amp; 135</td>
<td>160</td>
<td>200</td>
</tr>
<tr>
<td>Fuel</td>
<td>fuel oil # 5</td>
<td>LPG</td>
<td>LPG</td>
<td>LPG</td>
</tr>
<tr>
<td>Switching time, $t_s$ (seconds)</td>
<td>57, 67 &amp; 77</td>
<td>15, 20, ..., 60</td>
<td>30</td>
<td>10</td>
</tr>
<tr>
<td>$C_a/C_g$</td>
<td>14 – 19</td>
<td>6 – 16</td>
<td>2,5</td>
<td>10</td>
</tr>
<tr>
<td>$C_r/C_a$</td>
<td>0.9</td>
<td>0.9</td>
<td>0.68</td>
<td>1.13</td>
</tr>
</tbody>
</table>
5.2 Concepts for describing HiTAC features

Open-flame firing (direct heating systems)

1. Flame volume:

The determination of the flame volume, as a physical property of the flame in an industrial scale, was made by using the definition of the oxidation mixture ratio, $R_o$, which is a measure of the extent to which combustion at a given point in the flame has progressed [71]. This ratio can be calculated as follows [21]:

$$R_o = \frac{\sum c_c F_{c/o} \times R_o^{2}}{\sum c_c F_{F/c} \times R_o^{2}}$$  \hspace{1cm} (Eq. 1)

Where $S = n_o M_o / n_F M_F$, $\sigma$ is the mass fraction, $n$ is the stoichiometric coefficient (number of moles) and $M$ is the molecular weight. $R_o$ equals one at air inlet or when combustion is completed. However, it equals zero at fuel inlet. An empirical ratio of $R_o = 0.99$ has been assumed to indicate the outside border of the flame. Thus, the flame volume is confined to the region where

$$0 < R_o < 0.99$$  \hspace{1cm} (Eq. 2)

According to the above criteria, local flue gas composition measurements were utilized to identify the flame volume for the various configurations. The ratio between the flame and the furnace volume is referred to as the flame occupation coefficient, $R_F$.

2. Convective heat transfer coefficient determination:

Measurements of the total heat flux ($q_T$), i.e., the rate of heat flow per unit area on the surface located in the furnace, using the plug-type heat flux meter, were performed at different locations. In fact, this total heat flux meter served to complement the measured data obtained by the radiative heat flux meter, the irradiance, $G_r$, and gas temperature measurements, $T_g$, in order to obtain the convective heat transfer coefficient, $h_c$, in the interface between the meter surface and its environment. This is done by making an energy balance on the meter plug surface:

$$q_T = q_r + q_c = \left(\alpha_s G_r - \varepsilon \sigma T_s^4\right) + h_c \left(T_g - T_s\right) \hspace{1cm} (Eq. 3)$$

where

$q_c$ is the net convective heat flux received by the surface (kW/m$^2$),
$q_r$ is the net radiative heat flux between the surface and the hot environment, i.e. flue gases and furnace walls.
$\alpha_s$ is the absorptivity of the plug surface,
$\varepsilon$ is the emissivity of the plug surface,
$T_s$ is the temperature of the plug frontal surface.

Since the absorptivity $\alpha_s$ in equation (3) is nearly always equal to the emissivity $\varepsilon$ (gray surface) then the above equation can be written as:

$$q_T = \varepsilon \left(G_r - \sigma T_s^4\right) + h_c \left(T_g - T_s\right) \hspace{1cm} (Eq. 4)$$
The temperature of the plug frontal surface, $T_s$, can be calculated by knowing the thermal resistance of the plug and the temperature gradient thereof or by calibration curves. Thus, $h_t$ is the only variable left unknown in the above equation, and consequently $h_t$ and $q_c$ are calculated. However, since this method for determining $h_t$ involves several meters, it is not only costly and time consuming but also lacks accuracy (~25% uncertainty). Therefore, these investigations are only qualitative and were performed only on the first two configurations, the single-flame.

3. Spatial uniformity of measured parameter:

Furthermore, the spatial uniformity ratio, $R_{U_t}$, was used to calculate the uniformity of a temperature or a heat flux in a certain plane and was calculated according to the following equation:

$$R_{U_t} = \sqrt{\sum (U_i - \bar{U})^2}$$  \hspace{1cm} (Eq. 5)

where $U_i$ is the measured parameter time average for a complete cycle at a certain location or point and $\bar{U}$ is the arithmetic average of all measured points, $U$, in the plane.

4. Energy balance of the test furnace

In order to determine furnace efficiency for the three first operating configurations, energy balance for the test furnace was carried out. An energy system that included the test furnace and the regenerative burners was defined. The flow-rates and temperatures of all bulks entering and leaving the system boundary were measured to quantify all energy states and flows through the system. The energy balance equation for a system can be expressed as the rate equation of the 1st law of thermodynamics [72].

$$\dot{Q}_{sys} + \sum \dot{m}_i \left( h_i + \frac{u_i^2}{2} + gZ_i \right) = \frac{dE_{sys}}{dt} + \sum \dot{m}_o \left( h_o + \frac{u_o^2}{2} + gZ_o \right) + W_{sys}$$  \hspace{1cm} (Eq. 6)

The following assumptions were made in these calculations for simplification: (i) kinetic and potential energies were neglected; (ii) $(dE_{sys}/dt) = 0$ at periodic steady state conditions since logging data were made for a relatively long time; (iii) cooling air, fuel, and combustion air were all assumed to be moisture free and entering the system at the reference temperature, $T_R = 25^\circ C$; and (iv) the calculations were based on the lower heating value (LHV) and consequently the latent heat loss of vapor from chimney due to hydrogen content in fuel is not counted for.

The $\dot{Q}_{sys} = - \dot{Q}_{sur}$, i.e., energy losses through the furnace outer walls to the surroundings (surface losses). Therefore, the energy balance equation is reduced to

$$\sum \dot{m}_i \left( h_i \right) = \sum \dot{m}_o \left( h_o \right) + \dot{Q}_{sur}$$  \hspace{1cm} (Eq. 7)

The energy inputs, the left hand side of Eq. (7), in this case are

- chemical enthalpy in fuel, $\dot{Q}_F = \dot{\dot{m}}_F \cdot LHV$ ,
- physical enthalpy in fuel, $\dot{Q}_F$ ,
- sensible heat in combustion air, $\dot{Q}_a$ , and
The sensible heat or the physical enthalpy is,

\[ \dot{Q} = m \int_{t_i}^{t_f} C_p \, dT \]  

(Eq. 8)

The energy outputs, the first term in the right hand side of Eq. (7), are

- the sensible heat in cooling air outlet \( \dot{Q}_{c,o} \), and
- the sensible heat losses in the exhaust flue gas mixture, which is:

\[ \dot{Q}_{fg} = \dot{m}_{fg} \sum_{i} \int_{t_i}^{t_f} \alpha_i C_{p,i} \, dT \]  

(Eq. 9)

where \( \alpha_i \) is the mass fraction of each component \( i \). The net useful heating energy or the load of the furnace is \( \dot{Q}_{c,o} - \dot{Q}_{c,i} \). Other net energy outputs are the exhaust flue gas losses, Eq. (9), and surface heat losses by convection and radiation, \( \dot{Q}_{sur} \), which is measured by measuring the outer surface temperature of the furnace and the ambient temperature. The difference between the total energy inputs and the total energy outputs is considered as other losses. These other losses are probably due to the kinetic and potential energy losses that were ignored during these calculations or due to measurements inaccuracies.

Radiant tube (indirect firing systems)

In addition to the measurements of longitudinal and lateral temperature profiles, the heat flux incident on a surface from the tube was measured using the ellipsoidal meter in order to calculate the emissivity of the tube surface. Consequently, the net radiative heat flux, \( q \), from the tube was calculated from the following equation:

\[ q = \frac{\sigma}{A} \sum_i A_i \left( T_{AV,i}^4 - T_{AV,FR}^4 \right) \]  

(Eq. 10)

where \( i \) is one segment of the tube around every thermocouple and \( A_i \) is the segment area. Calculations were performed assuming various hypothetical average temperatures inside the furnace, \( T_{AV,FR} \) (e.g. 250°C, 500°C, 750°C, and minimum temperature, \( T_{AV,MIN} \), of the profiles, which always occurred in the case of recuperative system). The heat flux ratio, \( R_{HF} \), was then calculated using the equation below to permit a comparison between the two burner systems.

\[ R_{HF} = \frac{q_{RS}}{q_{CRS}} \]  

(Eq. 11)

Regenerative heat exchangers

The thermal performance of the heat exchangers is measured by means of the energy recovery ratio and the cold-side temperature efficiency (\( \eta \)). This efficiency is defined as \((T_{ao} - T_{ai})/(T_{g,r} - T_{ao})\). Since the combustion air temperature should always be above the self-ignition temperature of the fuel in
the application of HiTAC, $\eta_T$ is an important parameter because it is a measure of how close the combustion air temperature $T_{a,o}$ is to flue gas temperature $T_{g,i}$. The energy recovery ratio (ERR) is defined as $\dot{Q}_{g,i}/(\dot{Q}_{a,o} - \dot{Q}_{a,i})$, where the physical enthalpy, $\dot{Q}$, is calculated as per Eq. (8).

The effectiveness, $\varepsilon$, is:

$$\varepsilon = \frac{\dot{Q}}{\dot{Q}_{\text{max}}} = \frac{C_g (T_{g,i} - T_{g,o})}{C_{\min} (T_{g,i} - T_{a,i})} = \frac{C_o (T_{a,o} - T_{a,i})}{C_{\min} (T_{g,i} - T_{a,i})}$$  \hspace{1cm} (Eq. 12)

It is clear that $\varepsilon$ is equivalent to $\eta_T$ when $C_a < C_g$ in cases when the FSR is $\geq 100\%$ (or $C_o/C_g \leq 1$). In general, it is possible to express

$$\varepsilon = \frac{\int[N TU, C_a/C_g, C_r/C_o]}{\int}$$  \hspace{1cm} (Eq. 13)

The number of heat transfer units, $NTU$, is a non-dimensional expression of the heat transfer size of the exchanger. It serves as a measure for the quality of heat transfer between the gas and the matrix. The manner in which the heat transfer area and overall conductance enter into the $NTU$ is expressed as [49]:

$$NTU = \frac{A}{C_{\min}} \left[ \frac{1}{\left(\frac{1}{h_c} + h_c \right)_{g} + \left(\frac{1}{h_c} \right)_{o}} \right]$$  \hspace{1cm} (Eq. 14)

### 5.3 Experimental data evaluation methodology

**Dynamic response consideration**

Basically, measurements at all HiTAC configurations were carried out under periodic steady-state conditions, which is an unsteady state condition. Therefore, all raw measured data has been compensated for the inertia of the measuring device. This is in addition to the many measures, mentioned in the previous chapter, that were considered during measurements to cope with this dynamic operation. The compensated value was calculated as below [55].

$$I^{cm} = \tau \frac{dT_m}{dt} + T_m$$  \hspace{1cm} (Eq. 15)

Several measurements were performed to determine the time constant for several thermocouples and other measuring devices, e.g., on-line NOx analyser. However, the raw data from the thin-wire thermocouple were compensated to find the temperature fluctuation using the time constant value obtained from the manufacturer (25 ms).

In contrast, it is more challenging in the case of the total heat flux and gas composition measurements than in the case of other measurements in which only one sensor has to be considered, e.g., thermocouple. In the total heat flux meter, the temperature gradient is measured by two thermocouples along a steel plug. Methods for considering the unsteady-state condition effect on heat flux measurements have been also treated and used in this study [73]. Figure 13 shows the measured temperatures and the compensated temperature gradient of the total heat flux sensor.
Radiation effect on thermocouples

Another systematic error that should be considered, concerning temperature measurements, is the effect of thermal radiation exchange between thermocouples’ sheath and the hot environment in the furnace (e.g., furnace walls and gases). Therefore, thermal equilibrium cannot be achieved with the medium whose temperature is to be measured and result in measurement errors. Whenever a bare sheath of a thermocouple was inserted in the furnace to measure a flue gas stream temperature, energy balance was applied on the surface of the thermocouple in order to compensate the measured temperature for this effect.

![Figure 13. Measured, $T$, and compensated, $T^{\text{cm}}$, values of the cold and hot side thermocouples of THF meter and their compensated differential $\Delta T$.](image)

Measurement uncertainty

After estimation of these two systematic errors mentioned above and compensation of measured values, the uncertainties of final results were calculated according to Holman [74].

Uncertainties of species molar fraction measurements were between 2% and 3%. These values consider only the instrument and calibration gas accuracies and do not consider any other error source such as the introduction of probe or dissociation of some species.

Uncertainties for energy balance calculations of the furnace or the heat regenerator were around 3%, without considering errors generated due to compensation for dynamic or radiation effects. Judgement on these errors was left to the reader.

It is worthwhile to mention that the fuel used for all the above experiments, except for the fixed-bed, was a typical LPG used by the industry. According to the micro-GC measurement, it is composed mainly of 98% propane, 0.9% ethane, and 0.8% butane, with a lower heating value of 93.2 MJ/Nm$^3$. 
6. Theoretical Study

In addition to the experimental investigations mentioned in the previous chapter, theoretical investigations were required in order to achieve the first and second objectives set for this thesis. These are represented by thermodynamic investigations and developing mathematical models related to heat regenerators.

6.1 Thermodynamic aspects of OdC

The study approach of any new combustion technology begins with thermodynamic investigation. Thermodynamic analyses provide guidance to ideality and limitations. Enthalpy-temperature diagrams were constructed to investigate the temperature ranges and limitations at various Oxygen-diluted Combustion conditions, i.e., reactants at various oxygen mass fractions and temperatures. These diagrams, together with Temperature-Entropy diagrams, are shown in the Results chapter. In this section, 2nd law analyses were used on oxygen-diluted combustion to examine the possibilities for reducing irreversibility. Exergy and energy flows and losses were evaluated at every stream and stage in the combustion processes. A comparison was established between OdC systems utilizing different oxidizers, the HiTAC, and the flameless-oxy-fuel.

Approach

The oxygen-diluted combustion is a system comprising three components (dashed line in Figure 14) performing the processes of recuperating the heat of combustion product by the oxidizer, mixing of pre-heated oxidizer and combustion products to yield low oxygen concentration reactants, and the combustion of these reactants with the fuel. Although the low oxygen concentration is achieved in practice by internal flue gas recirculation, the recirculation and mixing process is hypothesized in a separate mixer located between the heat exchanger and the combustor as shown in Figure 14.

The following assumptions were also made in this analysis: (i) environment is dry air at 298.15K and 101.325 kpa; (ii) all processes are steady state steady flow; (iii) kinetic and potential energies are neglected; (iv) the oxidizer and the fuel enter the system at 298.15 K; (v) heat recuperation is a counter flow heat exchanger having an effectiveness of 0.9; (vi) dilution of oxidants is only due to gas recirculation (mixing of product gases with pre-heated or non-pre-heated oxidizers); (vii) the fuel is 100% propane; (viii) the oxidizer/fuel ratio was set to correspond to 3% oxygen in the combustion product gases (for complete combustion); (ix) combustion processes are adiabatic and isobaric; (x) assumed combustion product species are O₂, CO₂, H₂O, N₂, O, H, OH, H₂, CO, and NO; (xi) combustion gases entering the mixer or the heat exchanger are assumed to be a product of complete combustion containing only O₂, CO₂, H₂O, and N₂; (xii) irreversibility due to thermal interaction with environment is neglected in the mixer and the combustor. However, heat loss to the environment from a heat exchanger was accounted for based on some experimental data; (xiii) pressure loss is accounted for only in the heat exchanger; (xiv) constant pressure process in the mixer and combustor; (xv) thermodynamic gas properties obtained from data tables [7], [8], [75]; and (xvi) GASEQ code [76] used to calculate equilibrium states and temperatures.
Cases in which oxidizer is either air or oxygen were studied. In the case of conventional air OdC system (or HiTAC), a combined heat and gas-recirculation was considered. However, only gas-recirculation was considered in the case of a flameless-oxy-fuel system (oxy-fuel-OdC). Table 5 shows $R$ values and the corresponding oxygen mass fraction ($\varphi_{O2}$) of the reactants in the studied cases. Cases of oxy-fuel-OdD system with a combined gas and heat-recirculation will also be shown in the next chapter and compared with cases of only gas-recirculation.

Table 5. $R$-values and their corresponding oxygen mass fraction in the studied cases.

<table>
<thead>
<tr>
<th></th>
<th>$R$</th>
<th>0</th>
<th>0.5</th>
<th>1</th>
<th>2</th>
<th>4</th>
<th>6</th>
<th>100</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air</td>
<td>$\varphi_{O2}$</td>
<td>0.233</td>
<td>0.16</td>
<td>0.013</td>
<td>0.095</td>
<td>0.068</td>
<td>0.056</td>
<td>0.028</td>
</tr>
<tr>
<td>Oxygen</td>
<td>$\varphi_{O2}$</td>
<td>1</td>
<td>0.41</td>
<td>0.21</td>
<td>0.155</td>
<td>0.104</td>
<td>0.076</td>
<td>0.042</td>
</tr>
</tbody>
</table>

Irreversibility of the whole oxygen-diluted combustion system

The irreversibilities due to the three components in Figure 14 may or may not exceed that of an ordinary combustion. In order to examine that, $\dot{I}$ for mixing, heat exchange, and OdC processes were calculated.

**Combustor:** For an adiabatic combustion process, the irreversibility rate, $\dot{I}$ (in kW), can be calculated directly as

$$\dot{I} = T_e \dot{\Pi} \tag{Eq. 16}$$

where the entropy generation, $\dot{\Pi}$, is equivalent to $\Delta \dot{S}$ in this case. $\dot{I}$ can also be found from the exergy balance of the combustor.

$$\sum_{IN} \dot{E} - \sum_{OUT} \dot{E} = \dot{I} \tag{Eq. 17}$$

This is $\dot{E}_R + \dot{E}_F - \dot{E}_P = \dot{I}$, where the subscripts $R$, $F$, and $P$ refer to reactants, fuel, and products, respectively. The exergy rate of a flue gas stream of $m$ components is
\[
\dot{E} = \sum_{j} n_j \varepsilon_{ch,j} + \sum_{j} n_j \varepsilon_{ph,j} \quad (\text{Eq. 18})
\]

where \( n \) is the rate of number of moles. The specific molar chemical and physical exergies are

\[
\bar{\varepsilon}_{ch} = \sum_{j} x_j \bar{\varepsilon}_{ch,j} + \bar{R} T_e \sum_{j} x_j \ln(x_j) \quad (\text{Eq. 19})
\]

\[
\bar{\varepsilon}_{ph} = (T - T_e) \sum_{j} x_j \bar{\varepsilon}_{p,j} + \bar{R} T_e \ln\left(\frac{P}{P_e}\right) \quad (\text{Eq. 20})
\]

where the subscript \( e \) refers to the environmental condition, the superscript \( \sim \) indicates a molar quantity, \( x \) is the molar fraction, \( P \) is the total pressure of the mixture, and \( \varepsilon_p^e \) is the mean molar isobaric exergy capacity defined as

\[
\varepsilon_p^e = \frac{1}{T - T_e} \left[ \frac{\varepsilon_p dT}{T_e} - \frac{\bar{\varepsilon}_p^e dT}{T} \right] \quad (\text{Eq. 21})
\]

Since \( P = P_e \), the second term of the right hand side of equation (20) for the isobaric combustion process, is zero, it is useful to normalize \( \dot{I} \). Thus, the fraction that is lost from the total exergy input (\( \dot{E}_{ox} + \dot{E}_f \)) through irreversibility in the combustor is called the efficiency defect, \( \delta \).

**Heat exchanger:** In the calculation of \( \dot{I} \) for the heat exchanger, experimental values for pressure loss and heat loss to the environment were used and generalized for all cases. The total irreversibility is thus due to entropy change, Eq. (22), and the thermal interaction with the environment, \( \dot{I}^Q \) in Eq. (23). The later is equivalent to the exergy loss to the environment, \( \dot{E}^Q \).

\[
\dot{I} = T_e \left[ \dot{m}_{ox} \sum_{i} \left( T_{ox,i} \int_{T_{ox,i}}^{T_{ox}} x_i c_{pi} dT + R_{ox} T_e \ln\left(\frac{P_{ox,i}}{P_{ox,o}}\right)\right) \right. \\
\left. - \dot{m}_{fg} \sum_{i} \left( T_{fg,i} \int_{T_{fg,i}}^{T_{fg}} x_i c_{pi} dT + R_{fg} T_e \ln\left(\frac{P_{fg,i}}{P_{fg,o}}\right)\right) \right] \quad (\text{Eq. 22})
\]

\[
\dot{I}^Q = Q, \quad \frac{T_e - T_e}{T_e} = \dot{E}^Q \quad (\text{Eq. 23})
\]

The irreversibility or the efficiency defect due to the heat exchanger can also be found from the exergy balance as follows:

\[
\delta = 1 - \frac{\dot{E}_{ox,i} - \dot{E}_{ox,o}}{\dot{E}_{fg,o} - \dot{E}_{fg,i}} \quad (\text{Eq. 24})
\]
Mixer: \( I \) for the mixing

\[
I = T_v \left[ \dot{m}_{ao} c_{pa} \ln \left( \frac{T_v}{T_{ox}} \right) + \dot{m}_{fg} c_{ps} \ln \left( \frac{T_v}{T_{fg}} \right) \right] \quad \text{(Eq. 25)}
\]

2\textsuperscript{nd} law efficiency: For such multi component systems we can write the equation below to calculate, \( \psi \), the 2\textsuperscript{nd} law efficiency of the whole oxygen diluted combustion system.

\[
100\% = \psi + \delta_{HX} + \delta_{mix} + \delta_{comb} \quad \text{(Eq. 26)}
\]

where the last three terms in the above equation are the efficiency defect due to the heat exchanger, the mixer, and the combustor, respectively.

6.2 Mathematical modeling of heat regenerators

As mentioned earlier, there are two main types of ceramic heat storage materials: honeycomb and fixed bed. Geometry differences are not only in solid materials but also in gas flow channels. Therefore, two mathematical models were developed to numerically determine the dynamic temperature profiles of gases and solid storing materials: the first is a one-dimensional model for the fixed bed of spheres regenerator and the other is a two-dimensional model for honeycomb regenerator. The later takes into account the thermal conductivity parallel and perpendicular to the flow direction. It can simulate a two-composite material regenerator. Other advantages of these models are that all fluid and solid physical properties were set as a function of temperature and that the models are transient and can be used for burner operation configuration. Consequently, the thermal performance of the regenerator is evaluated at different matrix configuration and operation using these models. In addition, the models predict the pressure drop across the regenerator and other thermal parameters. The results are presented in a non-dimensional form in order to be a designing tool as well. These models, along with the experiments performed on the four heat regenerators, form a comprehensive experimental and theoretical study of the thermal performance of regenerators used in oxygen-diluted combustion burning systems, especially HiTAC.

Model for a fixed bed of randomly packed with spheres

A one-dimensional simulation model was developed [77]. This model is a simplification of the complicated geometry of the packing materials and the irregular flow that exists in the voids of the bed. Therefore, assumptions were made, e.g., \( \dot{u}_t \) is uniform packing, \( k_z \) is zero because the spheres contact at one point only, the size of the spheres is small compared to the bed size, and no heat loss through and within the regenerator from its boundaries, walls.

The governing equation is then formulated of the fluid, ignoring potential, kinetic, and viscous drag forces, and applied on both the solid flowing fluid phases, domains. The energy and mass balance equations of the fluid domain is:

\[
\rho C_p \frac{\partial T_g}{\partial t} - k \frac{\partial^2 T_g}{\partial z^2} + \rho C_p \dot{u}_z \frac{\partial T_g}{\partial z} = 0 \quad \text{(Eq. 27)}
\]

\[
\frac{\partial p}{\partial t} + \frac{\partial (\rho \dot{u})}{\partial z} = 0 \quad \text{(Eq. 28)}
\]
And the energy equation of the solid material domain is then [53]:

\[
\rho C_p \frac{\partial T_s}{\partial t} = k \frac{\partial}{\partial r} \left( r \frac{\partial T_s}{\partial r} \right) \tag{Eq. 29}
\]

Pressure drop across the regenerator can be determined using the Ergun equation [78], which is based on the ‘tube bundle model’ approach.

\[
\Delta P = \left[ 150 \left( \frac{1 - \phi}{d_p \cdot G / \mu} \right) + \frac{7}{4} \right] \cdot \left[ \frac{G^2 (1 - \phi)}{\rho \phi^3 d_p / L} \right] \tag{Eq. 30}
\]

Figure 15. The fixed bed and the periodic boundary conditions.

Model for honeycomb regenerator

Because of the geometric symmetry of the honeycomb structure, a two-dimensional simulation model was developed (see supplement II) for one honeycomb cell, or matrix, which formed a small part of the regenerator cross-section along the flow path (Figure 6). The regenerator can also be composed of two different materials along the heat exchangers, e.g. alumina (99.7% pure \( \text{Al}_2\text{O}_3 \)) and cordierite (Figure 16).

The energy equation of the solid material domain is:

\[
\rho C_p \frac{\partial T_s}{\partial t} = k \nabla^2 T_s \tag{Eq. 31}
\]

And the energy equation of the flowing gases domain is:

\[
\rho C_p \frac{\partial T_g}{\partial t} + \nabla \cdot ( -k \nabla T_g + \rho C_p T_g u ) = 0 \tag{Eq. 32}
\]

The above equation is coupled to the momentum and mass balance equations in order to obtain the velocities \( u_x \) and \( u_y \). The flow is laminar in the honeycomb and the equations for momentum, Navier-Stokes, and mass balances are:
\[
\frac{\partial u}{\partial t} - \mu \nabla^2 u + \rho(u, \nabla)u + \nabla p = 0 \quad \text{(Eq. 33)}
\]

\[
\nabla \cdot (\rho u) = 0 \quad \text{(Eq. 34)}
\]

For such simple geometry with very small \(D_h\) (normal size is attributed to 100 cell/in\(^2\)), the flow is always laminar since local \(Re\) ranges between ~90 and 240 at nominal operating conditions for the 100 cells/in\(^2\) honeycomb. Therefore, the velocity profile and pressure drop across the regenerator can be determined from the solution of Navier-Stokes equation,

\[
u_x = 4u_{max} \cdot j(1-j) \quad \text{and} \quad u_y = 0 \quad \text{(Eq. 35)}
\]

\[
\Delta P = 32\mu(u) \frac{L}{D_h} \quad \text{(Eq. 36)}
\]

where \(j\) in Eq. (35) runs between 0 and 1 between two-opposite walls of a cell in the direction perpendicular to the flow, and \(u_{max} = 2u_{in}\), \(u_y\) can be found at any cross section by knowing the gas inlet velocity and the ideal gas law. Replacing Eq. (33) by Eq. (35) and (36) will minimize the number of computational steps, without sacrificing the accuracy of the results.

**Boundary conditions and closure**

The boundary conditions for such transient problems are periodic, as shown in Figure 15 and Figure 16. They change every time the burners switch over (\(t_s\)). BC1 is in the interface between the two domains, \(h(T_s-T_g)\), and couples Eq. (27) with Eq. (29) in the case of the fixed bed model and Eq. (31) with Eq. (32) in the case of honeycomb. It is for the flow domain and a negative sign is added for the solid domain. \(h\) includes the convective and radiative heat transfer coefficient, \(h_c\) and \(h_r\).

![Figure 16. Cross-section of one cell showing the boundary condition numbering, flow direction before and after switching and unknown parameters in every domain.](image)

The governing equations for the fixed bed model Eq. (27, 28, and 29) and their boundary conditions involve six unknowns. The difference between the number of unknowns and equations is also three in the case of the honeycomb model. Thus, the following three equations are required to close the problem; the ideal gas law for the gas-phase state equation and equations for \(h_c\) and \(h_r\). \(hr\) is due to radiation emitted from gases and absorbed by walls during the regeneration mode. \(h_c\) was obtained using correlation attributed to Coppage, London, and Denton [48] in the case of fixed bed Eq. (37) and using the formula attributed to Hausen [79], [80] in the case of honeycomb, Eq. (38).
\[ \text{St} \cdot \text{Pr}^{2/3} = 0.23 \text{Re}^{-0.3} \quad \text{(Eq. 37)} \]

\[ \frac{h \cdot D_h}{k_g} = \frac{Nu + 0.0668 (D_h/L) Re Pr}{1 + 0.04 (D_h/L) Re Pr^{2/3}} \quad \text{(Eq. 38)} \]

where \( Nu \) is the nusselt number that is constant and dependent on the geometry of the flow cross section and \( D_h \) is of a cross section perpendicular to the flow direction.

**Computation procedure**

In each model, governing equations of the two domains become a set of coupled nonlinear equations that are solved numerically. FEMLAB® [81], modeling software for finite-element analysis of coupled partial differential equations, was used for numerical integration of these equations to solve for the temperature profiles and for the pressure drop across the regenerator. The transient solver in FEMLAB is an implicit time-stepping scheme, which implies that it must solve a non-linear system of equations at each time step. This software code is currently called as ‘COMSL Multiphysics™’.

The computations are initiated using cold start initial conditions and executed for one complete cycle of operation, i.e., the duration for both the regeneration and the combustion periods. The results are then saved in the computer and the final temperatures are set to be the initial conditions of the next run. The computations were executed repeatedly until the temperature profiles just before the change-over were the same as those of the previous data, a periodic steady state. The regenerators were simulated at several boundary and operating conditions.

The average viscosity, \( \mu \), and density, \( \rho \), in Eq. (30) were determined by integrating their values along the regenerator at every time step.
7. Results

7.1 Energy balance measurements

Energy balance calculations for both conventional and HiTAC configurations are shown in Figure 17. The efficiency of the furnace resulted by single-flame and twin-flame HiTAC was higher than that resulted by conventional flame, by 35% and 44%, respectively. The term ‘uncounted losses’ represents those such as Kinetic and Potential energies for the masses entering and leaving the system boundary or due to measurement inaccuracies.

It was apparent that air pre-heating increases the heating potential of the furnace gases, i.e., the useful energy [34]. The fuel saving by high pre-heating air combustion is larger than that of the saving corresponding to the heat recirculation. For example, the heating potential, represented by heating the air, is higher by more than three times in the case of twin-flame HiTAC compared to conventional-air combustion.

![Figure 17. Net heat output of test furnace resulted by both conventional and HiTAC flame.](image)

7.2 Thermodynamic analysis of Oxygen-diluted Combustion

Enthalpy – Temperature diagram of combustion process

Conventional thermodynamics analysis was made to construct the H-T diagram shown in Figure 18a. The $H_R$ curve is the enthalpy of the unburned reactants as a function of temperature for a mixture of fuel and air ($\omega_{O2} = 0.23$). The other curves, underneath and parallel to it, were also the enthalpies of unburned reactants but for diluted air and fuel. The $H_P$ curve is the enthalpy of a chemically equilibrated burned product. It is independent of oxygen concentration because the products of combustion at equilibrium and at a certain temperature are the same for all cases if dilution is made by flue gas-recirculation, as assumed in this study. Since the two curves ($H_R$ and $H_P$) are converging due to the thermal dissociation and the increase in specific heat of the product, heat of combustion becomes less until it becomes zero, which corresponds to the adiabatic limit temperature $T_{alf}$. It is clear from the figure that $T_{alf}$ decreases with dilution increase. $T_{alf}$ decreases from 3233 K to 2520 K when the oxygen mass fraction in the reactant is reduced from 23% to 5.6%. Figure 18b shows the same $H-T$ diagram but in the case of oxy-fuel-OdC system.

As seen from Figure 18a, if conventional air is pre-heated to 1200K, the temperature increase due to adiabatic combustion is about 1400 K (follow point a to b). However, if this pre-heated air contains only 5.5% oxygen by mass (point c), then this temperature increase would be only ~300 K (follow point c to d). It is evident from these graphs that the temperature increase is thermodynamically limited and bounded when the oxygen concentration of the reactants is reduced.
Entropy generation

Depending on the value of $R$, the oxygen-diluted combustion involves variations of mass flow rates for the same energy input of the fuel. Therefore, it is more convenient to use the absolute entropy rate $\dot{S}$ (J/K.s) to compare oxygen-diluted combustion with other combustion processes. By comparing curve I with II and III in the T-S diagram (Figure 19), the entropy generation for an adiabatic combustion process is reduced to more than 60% due to the effect of either pre-heating or oxygen enrichment. The calculations made in this example are based on 200 kW energy input from propane. In the case of pre-heating the reactants, the temperature increase in the product gases is less than that in the reactant gases because of the increase of specific heat caused by thermal dissociation in product. As a result, less entropy is generated due to pre-heating. In the case of oxygen combustion, although $\Delta T$ is higher than that of air combustion, entropy generation of oxygen combustion is minimized because N$_2$ dilution is reduced and thus reduces the heat capacity of both reactants and products [8].
Figure 19. Effect of preheating of the reactants and oxygen enrichment on the entropy rate generation in an adiabatic isobaric combustion process.

Figure 20. Effect of dilution by flue gas recirculation on the entropy rate generation in an adiabatic isobaric combustion process.

Figure 20 shows the effect of the gas-recirculation on entropy generation for both conventional-OdC and oxy-fuel-OdC systems. In both cases, R was set to 6 and 35, respectively, by which O2 concentration was reduced to about 5%. By comparing curves I and II from Figure 20 with curves II and III from Figure 19, one can conclude that entropy generation is increased by about 14–18 % due to gas-recirculation. However, and in the case of oxygen, pre-heating oxygen before mixing will further minimize $\Delta S$ (comparing curves II and III in Figure 20). Figure 19 and Figure 20 were made in the same scale to clearly show that comparison based on the maximum temperature of the products is in favor of gas-recirculation.

Irreversibility of Oxygen-diluted Combustion process

Figure 21 shows the relation between the efficiency defect due to combustion, $\delta_{\text{comb}}$, and the temperature of the combustion gas products. It is clear that we can get higher 2nd law efficiency if oxygen concentration in the reactants is reduced and/or if pre-heating raises the combustion products temperature. And therefore, the combination between heat and gas-recirculation produces the least irreversibility.
Although there is no heat exchanger (oxidizer pre-heater) in the case of an oxy-fuel-OdC system (Figure 21b) in which mixing only performs the pre-heating of the reactants, the efficiency defect is slightly lower in the case of an oxy-fuel-OdC system than that of the case of an air-ODC system if we keep the products temperature constant (see the dotted vertical lines at 2000 K in Figure 21a and Figure 21b).

Figure 21. Efficiency defect due to combustion as a function of equilibrated temperature of the products for various oxygen concentration levels.

Irreversibility of the whole Oxygen-diluted Combustion system

Figure 22 shows the particular efficiency defect of every component of the system at relatively high gas-recirculation rates. It should be noted again that the heat exchanger is absent in the oxy-fuel-OdC system and therefore maximum temperatures are not as high as in the case of a conventional-OdC system. Since mixing in the case of air takes place at lower temperature differences compared to that of oxygen, $\delta_{\text{mix}}$ in the oxygen case is much higher than that of the air cases, and it can be even higher than both efficiency defects $\delta_{\text{HX}}$ and $\delta_{\text{mix}}$ in the case of air. Therefore, the total efficiency of both systems is almost comparable. Point (P) in Figure 22b, in contrast, represents the total
efficiency defect of an Oxygen-diluted Combustion when both gas and heat-recirculation is used in the case of oxy-fuel-OdC system. It is obvious that adding heat-recirculation increases the total efficiency of the system.

![efficiency defect graph](image)

**Figure 22. The efficiency defect of every component of the oxygen-diluted combustion system as a function of equilibrated temperature of the product combustion gases.**

The bar chart in Figure 23 shows a comparison between an ordinary combustion process (the first column) and oxygen diluted ones at low oxygen concentrations (bars 2, 3, and 4). The second column is for a conventional air-OdC system, while columns 3 and 4 are for an oxy-fuel-OdC system. In column 3 only gas-recirculation takes place, while in column 4, gas and heat recirculation are taking place. It is obvious from this comparison that the oxygen system is the most efficient system compared to others, especially when adding the heat exchanger. The irreversibilities due to the heat exchanger and the mixer together (column 4) are less than that of the mixer alone (column 3) when mixing performs the pre-heating. This additional pre-heating using the heat exchanger (heat recirculation) increases the reactant temperatures, and thus further reduces the irreversibility in the combustion process. The overall increase of efficiency is about 8% when adding the heat exchanger to the oxy-fuel-OdC system.
The last three columns (5, 6, and 7) in Figure 23 are for cases of extreme gas-recirculation ($R = 100$) to show the trend for high 2nd law efficiency of the oxygen-diluted combustion process. It is clear that $\delta_{\text{comb}}$ is reduced with gas and heat-recirculation increase. Although $\delta_{\text{mix}}$ and $\delta_{\text{HX}}$ increases when $\omega_{O2}$ of the reactants decreases, the 2nd low efficiency of the whole OdC system increases and is higher if oxygen is being used as an oxidizer. In fact, if the same trend is followed, not only the 2nd law efficiency increases but also the first law efficiency increases. Flue gas energy loss of oxygen OdC system is less than 3% of the total energy in the fuel if a recuperator of 0.9 effectiveness is used to pre-heat oxygen (the cases of columns 4 and 7). This loss is about 10% for an air OCD system (columns 2 and 5).

![Figure 23. Comparison between an ordinary combustion and various cases of oxygen-diluted combustion.](image)

7.3 Performance of the heat regenerators used in HiTAC burning systems.

Mathematical modelling

Temperatures, pressures, and gas velocities can be determined at any location along the regenerator and at any time [77, supplement II]. Figure 24 is a plot of the time change of temperatures from the cold start up of the regenerator of the honeycomb regenerator, reg. no. 2, at its cold and hot ends. The switching time is 30 seconds and it takes only two cycles, i.e., two minutes, to reach the required combustion air temperature for HiTAC process, although it requires 11 cycles to reach the periodic steady state operating condition. In the case of the fixed bed, periodic steady state operation is reached after three hours, and temperatures become suitable for HiTAC after 30 minutes.

On the other hand, Figure 25 is a plot of the time change of parameters for the fixed bed regenerator, reg. no. 1, but after the periodic steady state is reached. The flue gas and combustion air exit temperatures can be determined from the cold and hot ends temperatures curves, respectively in this figure. The knowledge of such temperatures is significant to the calculation of the regenerator thermal performance. Figure 25 also shows the gauge pressure reading located at the cold end of the
regenerator. In the calculation of this value, the pressure at the hot end, the furnace, is assumed to be zero. Therefore, the pressure difference due to a twin regenerators in operation would be the positive pressure of combustion air at a certain time minus the negative pressure of flue gases 67 seconds earlier or after, i.e. exactly equivalent to the case when both regenerators are simulated simultaneously. At periodic steady state, this pressure drop was ~ 315 mmH₂O.

Figure 24. Honeycomb reg. #2: time history of ends inlet and exit temperatures at x = 0 and x = L.

Pressure drop across the honeycomb regenerator at periodic steady state was only 1.16 kpa for regenerator #2, compared to 4.75 kpa in the case of a fixed bed regenerator having an equivalent NTU value.

Figure 25. Fixed bed reg. #1: Time history of ends temperatures and gauge pressure after periodic steady state.

Moreover, the numerical calculation can also predict the temperature profiles of the solid storing material. Radial temperature profiles of the solid spheres in the case of the fixed bed are shown in Figure 26a and in Figure 26b during regeneration and combustion periods, respectively. The sphere is located at the hot end, z = 0, and the switching time is 67 seconds. Temperature profiles are plotted at times mentioned in the graphs.
Figure 26. Radial temperature profiles of a spherical solid particle, 14mm diameter, at several times during (a) regeneration and (b) combustion periods.

Figure 27 shows the thermal performance of the fixed bed regenerator made for regenerator # 1 and the honeycomb regenerator, reg. # 2, according to the mathematical simulation results. The results of each regenerator are contained inside a dotted line envelope. Although the two heat exchangers are so different in type and geometry, they are identical in terms of \( \frac{C_r}{C_g} \) and therefore can be presented in one graph in a dimensionless form. In the case fixed bed model, simulations were carried out for different bed and sphere sizes corresponding to these NTU values.

Figure 27. Simulation results of the two models, ineffectiveness as a function of NTU and matrix capacity-rate ratio for the fixed bed and honeycomb regenerators.

Logically, the effectiveness of identical \( C_r/C_g \) and \( NTU \) values in both regenerators should be similar. However, if comparison is made between the results of two models (say at \( C_r/C_g = 16 \)), differences of about 2-3% will be found in the effectiveness value for the same \( NTU \) value. This
difference is mainly because the thermal conductivity parallel to the flow direction was considered only in the two-dimensional model (the honeycomb model), but not in the one-dimensional model (the fixed bed model). Differences in accuracies between the two models also contribute in these differences.

Experimental results

Figure 28 shows the results of experiments carried out on four different regenerators to examine their thermal performance and to verify the mathematical models. The number of different operating conditions of these experiments is limited by the capabilities of the burners, especially in the fixed bed regenerator case. However, verification of the above model is still possible and both models and experiments show a very good agreement, especially at short switching times, i.e., high $C_r/C_a$. Differences are between 1% and 3% and may reach 5% at long switching times operation. The reason is because in reality there are other components in the regenerative burning system that play the role of additional regeneration material as the fluid passes through during operation. Normally these components have higher mass and storage capacitance than the honeycomb, and that is why they are capable to store and recover energy at longer switching times. The uncertainties of these experimental effectiveness and ERR were 3% and 4%, respectively.

Regenerator #3 and #4 are other honeycomb regenerators that have values of $C_a/C_g$ different than that of regenerator #2, where $C_a/C_g$ is 0.9. When comparing regenerators at similar NTU values but different $C_a/C_g$ values, it is clear that effectiveness increases with a $C_a/C_g$ decrease (Figure 28). However, Figure 29 shows that the ERR is highest when $C_a/C_g$ is 0.9, i.e., when all combustion gases passes through the regenerator again (FSR=100%). Note also that the cold side efficiency is equivalent to the effectiveness of the regenerators with $C_a/C_g < 1$. However, supplying the combustion air at a high temperature required for the HiTAC process can be crucial in the case of regenerators with $C_a/C_g > 1$ because the cold side temperature efficiency is normally low for these regenerators.

Figure 28. Experimental results, ineffectiveness as a function of NTU, and matrix capacity-rate ratio for the four tested regenerators.
Figure 29. Thermal performance parameters, $\varepsilon$, ERR, $\eta_T$ for some commercial regenerators as a function of $C_a/C_g$.

7.4 Local flue gas compositions inside the test furnace

Figure 30 and Figure 31 (see also supplement III) present the flue gas compositions at various points inside the semi industrial test furnace resulted by the first two studied configurations, namely single-flame conventional and single-flame HiTAC (shown in Figure 8a and b, respectively). The measured locations are shown with points.

Figure 30. Vertical planes inside the furnace show flue gas molar fractions as a result of single-flame conventional-air combustion configuration. Coordinates are in Y-Z Plane, X=0.
It is apparent from these measurements that:

− the chemistry field is more uniform in the HiTAC configuration and no high gradients of species concentration were found;

− the propane was not detected at all when operating at conventional-air configuration, while it was detectable downstream to 1.2 m from the burner (Figure 31) in the HiTAC flame. Therefore, the high speed air jets seem to penetrate deeply into the furnace; and

− the reaction zone for the HiTAC flame is larger than that of the conventional flame because more hydrocarbons were detected in broader regions in the furnace and at longer distances downstream from the burner nozzles. In single-flame conventional combustion (Figure 30), only hydrogen, carbon monoxide, and up to 500 ppm butane was detected 300 mm downstream from the burner center. However, in the single-flame HiTAC (Figure 31), a considerable amount of combustibles were detected in a wider region in the furnace. For example, the CO molar fraction in HiTAC (Figure 31) has a peak of 2.45% at 0.6 m from the burner. While in the conventional flame (Figure 30), the peak is 0.6% and at shorter distance from the burner. Almost similar profiles of H₂ were also detected in the cases of HiTAC. The presence of hydrogen indicates the slow combustion as well as cracking of the fuel [31].

This gradual disappearance of the fuel and the appearance of the intermediate species hydrocarbons in relatively high concentrations and at broader regions inside the furnace in the case of HiTAC is evidence of a larger reaction zone (or flame) and of a high concentration of radicals that are responsible of the final burn out of these species.

In addition, measurements were carried out for another HiTAC-based configuration, namely the twin-flame counter configuration (Figure 8c). The first three graphs in Figure 32 shows the molar fraction distribution for C₂H₆ and C₂H₂, CO, and H₂ for this configuration. The existence of these components is also an indication for a large and uniform reaction zone in a case of twin-flame HiTAC. For this configuration, however, presenting the local measurements is challenging. This is because the number of openings in the roof of the furnace that are made for investigations and probes insertions is limited to 13 due to construction limitation. Moreover, and as shown in Figure 5, distances between measuring points and a pair of burners (A and C) are not similar to that of the other pair (B and D). Nevertheless, the mesh of the measuring points for twin-flame burners can be increased by more than double by a data reshuffling method by which the measuring data is obtained, when a pair of burners is in combustion, can be shifted to that of the other pair when they are in combustion too. The results of this method for CO, H₂, and O₂ molar fraction distribution, resulting when burners B and D are in combustion mode, are shown in the last three graphs of
Figure 32. The arrows in the first graph show the flow directions of entering combustion air and exiting flue gases when burners B and D are in combustion mode. Unlike the coordinate system used in the single-flame configurations, burner B was chosen to be the origin (0, 0, 0) in presentation of these graphs.

Figure 32. Horizontal planes inside the furnace showing flue gas molar fractions as a result of the twin-flame counter HiTAC configuration. Coordinates are in X-Z Plane, Y=100.
7.5 Chemical flame volume

It was assumed that the border of the flame is at $R_o \sim 0.99$. According to this category, only hydrogen and carbon monoxide were found as combustibles in the flame front. For example, a mass fraction of 18 ppm and 580 ppm of $H_2$ and CO, respectively, can satisfy this category (this corresponds to a measured 300 and 700 ppm molar fraction of $H_2$ and CO, respectively, dry basis). Therefore, the flames borders were identified. Figure 33 and Table 6 show a comparison of flame geometry for the first three configurations. Single-flame and twin-flame HiTAC were larger than the conventional flame by 37 and 25, respectively; this corresponds to around 4% and 2.5% of the furnace space. However, calculation of $R_o$ close to the burner nozzles, in the case of the twin-flame burner, was not possible because no measurement had been carried out in that region.

![Conventional flame](image)

![HiTAC twin-flame](image)

**Figure 33. Flame shape and geometry inside the test furnace for the first three configurations.**

**Table 6. List of flame characteristic for the three studied cases.**

<table>
<thead>
<tr>
<th>Combustion mode</th>
<th>Flame volume (m3)</th>
<th>Flame length (mm)</th>
<th>Maximum flame diameter (mm)</th>
<th>Mean flame diameter (mm)</th>
<th>$R_o$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conventional flame</td>
<td>0.0072</td>
<td>872</td>
<td>161</td>
<td>113</td>
<td>0.1%</td>
</tr>
<tr>
<td>Single-flame HiTAC</td>
<td>0.2706</td>
<td>2803</td>
<td>495</td>
<td>328</td>
<td>3.7%</td>
</tr>
<tr>
<td>Twin-flame HiTAC</td>
<td>0.1836</td>
<td>1342</td>
<td>525</td>
<td>390</td>
<td>2.5%</td>
</tr>
</tbody>
</table>

7.6 Temperature profiles

Figure 34 shows the measured temperature profiles of single-flame combustion configurations (a and b) using the pyrometer. The half plane to the left is for a HiTAC flame (configuration b in Figure 8) and the other half is for conventional air combustion (configuration a). The measured locations are shown with points. In spite of the dynamic operation of switching devises and the regenerators, time variation of the measured temperatures were not observed in all points and the profiles shown are the time average temperature profiles.
However, this situation was different in the case of twin-flame configurations. Therefore, a thin-wire thermocouple was used to measure the instantaneous temperature at various locations. Figure 35 shows the temperature profiles of the HiTAC twin-flame system for two configurations: counter and parallel (configuration c and d in Figure 8). The profiles are shown only for one unit (burner pair B and D) and the dimensions are relative to burner B. The profile to the left of the burner is for the average temperature during the combustion period of burner B, while the one to the right is for the average temperature during the regenerative period of burner B. The temperature profiles of a stagger configuration, not shown, were very similar to the parallel configuration, probably because the flame in both configurations has straight axis.

![Figure 34. Temperature profiles of single-flame HiTAC and conventional-air combustion in planes at burner center.](image)

![Figure 35. Temperature profile of a twin-flame in a plane at burner center.](image)

The spatial temperature uniformity ratio, \( R_{U} \), was found to be 1.1, 0.33, and 0.23 for the cases of conventional single-flame HiTAC single-flame, and HiTAC twin-flame counter, respectively. It is evidenced from the above two figures that:

- the spatial temperature distributions of all HiTAC configurations are more uniform than that of a conventional air combustion;
- the time variations of these profiles are also uniform in all cases except for the twin-flame counter configuration; and
- in spite of the high temperature of the combustion air in all HiTAC configurations, the peak temperatures of the HiTAC flames are significantly low and are lower than that of the conventional-air combustion.

Thermodynamically, the temperature increase due to combustion is mild when the air and fuel jets are diluted first. (See Figure 18a and Supplement II.)

The total error of the measurement was found to be 27°C.

**Temperature fluctuation maps**

Figure 36 presents these fluctuations for two twin-flame configurations and shows small fluctuations in the measured points of about one order of magnitude compared to that of other
conventional-air flames of [2], [12], and [82]. Moreover, these fluctuations are smaller when the flame axis is straight, i.e., in parallel and in staggered configurations.

The profiles of fluctuation in temperature are relatively even and the highest fluctuation is in the region closest to the burner exit (opening 6 in Figure 5).

Gupta et al. [82] indicated that when the fluctuation is small the integral and micro thermal time scales are high, and therefore the turbulence intensity in the flame is low.

7.7 Heat flux measurements - thermal radiation and convection

Figure 37 shows the measured thermal radiation incidence, $G_{rr}$, on the ceiling of the test furnace for the five configurations using the ellipsoidal radiometer. These are time average values with 8 kW/m² uncertainty. Local total heat flux, $q_T$, values were measured to calculate $h_c$. (Results can be seen in Supplement IV). Before discussing the results of these heat transfer measurements, the following facts should be considered:

1- Although all tests were performed under the same firing capacity and furnace wall temperature, the heat carried out by the air-cooling tubes, the heat sink, were more than two and a half to three times higher in the HiTAC cases. Consequently, the tubes’ outer surface temperatures were lower and hence affected and reduced the radiation incident on the meter. Therefore, the overall impression about the heat transfer to a heat sink is much higher in the case of HiTAC and cannot be judged by the sole reading of this meter.

2- The measurement of convective heat transfer is just qualitative and flow conditions around the meter surface do not prevail a real object or slab to be heated.

Accordingly, measurements prove that in the twin-flame configuration the time variation of the radiative heat flux due to switching was between 5% and 10%, varying from point to point. The highest and most uniform radiation was in the case of HiTAC single flame. The uniformity ratios, $R_t$, were found to be 0.28, 0.18, 0.26, 0.25, and 0.18 for the conventional, single HiTAC, counter, parallel, and stagger configurations, respectively. It is obvious that values of $h_c$ at lateral distances of 400 mm away from the burner axis, and further, are uniform and in the range of 70 W/m²K for both combustion configurations. For the HiTAC case, this corresponds to ~60 kW/m² net
convective heat flux, \( q_c \), to the meter surface compared to \( \sim 135 \text{ kW} \) net radiative heat flux, \( q_r \), between the meter plug surface and the hot environment. Thus, the convective heat transfer accounts for about 30% of the total heat transfer.

Figure 37. Thermal radiation incidence on the ceiling (kW/m\(^2\)) (a) conventional air (b) single-flame (c) twin-flame counter (d) twin-flame parallel (e) twin-flame stagger.
7.8 Effective radiation from a radiant-tube based on HiTAC

Supplement V provides more detailed results about the radiant-tube experiments. Comparison between a recuperative system, the configuration of Figure 8f, and a HiTAC-based regenerative system, the configuration of Figure 8g, at different operating conditions has been carried out experimentally. The results are summarized as follows:

- Longitudinal temperature profiles were more uniform in the case of a regenerative system partly because of the cyclical nature of the firing process with twin burners, one at each end, and partly because of the oxygen diluted combustion process, i.e., large flame volumes and small temperature gradient in the flames.

- The longitudinal temperature profiles uniformity caused the cross sectional temperature profiles, around the cross section of the tube, to be more uniform, as well in the case of the regenerative system, as compared to the conventional recuperative system. In the case of the recuperative system the temperature difference between the top and bottom side of the tube is higher (~25°C). This difference is due to uneven heating for the tube with the recuperative system.

- The temperature levels are much higher along the radiant tube for the regenerative system, corresponding to a higher heat flux potential for this system. Therefore, the net radiative heat flux from the tube is always higher for the regenerative system, as shown in Figure 38 and Table 7. The difference between the systems also depends on the operating conditions, including the average temperature in the furnace, $T_{AV,FR}$. At high temperatures the radiative heat flux from the tube is about twice that of the conventional recuperative system, resulting in a higher radiative heat flux ratio, $R_{HF}$.

<table>
<thead>
<tr>
<th>No.</th>
<th>Test no.</th>
<th>$T_{AV,FR}$ (250°C)</th>
<th>$T_{AV,FR}$ (500°C)</th>
<th>$T_{AV,FR}$ (750°C)</th>
<th>$T_{AV,FR}$ ($T_{AV,MIN}$)*</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>q</td>
<td>$R_{HF}$</td>
<td>$Q$</td>
<td>$R_{HF}$</td>
</tr>
<tr>
<td>-</td>
<td>-</td>
<td>kW/m²</td>
<td>1.36</td>
<td>kW/m²</td>
<td>21.6</td>
</tr>
<tr>
<td>I</td>
<td>CRS-1</td>
<td>31.8</td>
<td>43.4</td>
<td>-</td>
<td>21.6</td>
</tr>
<tr>
<td></td>
<td>RS-1</td>
<td>42.8</td>
<td>49.7</td>
<td>-</td>
<td>32.6</td>
</tr>
<tr>
<td>II</td>
<td>CRS-2</td>
<td>42.8</td>
<td>49.7</td>
<td>-</td>
<td>32.6</td>
</tr>
<tr>
<td></td>
<td>RS-2</td>
<td>42.8</td>
<td>49.7</td>
<td>-</td>
<td>32.6</td>
</tr>
<tr>
<td>III</td>
<td>CRS-3</td>
<td>60.1</td>
<td>67.3</td>
<td>-</td>
<td>49.8</td>
</tr>
<tr>
<td></td>
<td>RS-3</td>
<td>60.1</td>
<td>67.3</td>
<td>-</td>
<td>49.8</td>
</tr>
<tr>
<td>IV</td>
<td>CRS-4</td>
<td>77.6</td>
<td>82.3</td>
<td>-</td>
<td>67.4</td>
</tr>
<tr>
<td></td>
<td>RS-4</td>
<td>77.6</td>
<td>82.3</td>
<td>-</td>
<td>67.4</td>
</tr>
</tbody>
</table>

* Values of $T_{AV,MIN}$ are shown in Figure 38.
- The efficiency of the whole system is higher for the regenerative system cases of up to 25%, compared to conventional recuperative system (Table 8). The improved efficiency with the regenerative system becomes more significant with higher reference point temperatures. This higher efficiency is mainly due to the lower temperature of the flue gases and the lower surface losses, because of the smaller surface area of the burner units and pipes in comparison with the recuperative system.

Table 8. Flue gas temperatures, surface and flue gas losses and efficiency of the system.

<table>
<thead>
<tr>
<th>No.</th>
<th>Test no.</th>
<th>(T_{fl})</th>
<th>(\dot{Q}_{fl})</th>
<th>(\dot{Q}_{sur})</th>
<th>(\eta)</th>
<th>(\Delta\eta)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>°C</td>
<td>kW</td>
<td>kW</td>
<td>%</td>
<td>%</td>
</tr>
<tr>
<td>I</td>
<td>CRS-1</td>
<td>548</td>
<td>19.6</td>
<td>3.3</td>
<td>71.6</td>
<td>13.4</td>
</tr>
<tr>
<td></td>
<td>RS-1</td>
<td>261</td>
<td>8.8</td>
<td>2.5</td>
<td>85.1</td>
<td></td>
</tr>
<tr>
<td>II</td>
<td>CRS-2</td>
<td>678</td>
<td>40.4</td>
<td>4.5</td>
<td>65.8</td>
<td>17.5</td>
</tr>
<tr>
<td></td>
<td>RS-2</td>
<td>309</td>
<td>16.6</td>
<td>2.9</td>
<td>83.3</td>
<td></td>
</tr>
<tr>
<td>III</td>
<td>CRS-3</td>
<td>730</td>
<td>44.0</td>
<td>5.3</td>
<td>62.9</td>
<td>21.1</td>
</tr>
<tr>
<td></td>
<td>RS-3</td>
<td>298</td>
<td>16.4</td>
<td>3.6</td>
<td>84.0</td>
<td></td>
</tr>
<tr>
<td>IV</td>
<td>CRS-4</td>
<td>815</td>
<td>58.9</td>
<td>6.1</td>
<td>58.6</td>
<td>24.2</td>
</tr>
<tr>
<td></td>
<td>RS-4</td>
<td>325</td>
<td>21.4</td>
<td>4.1</td>
<td>82.8</td>
<td></td>
</tr>
</tbody>
</table>

In practice, it can be concluded that one of the advantages of the application of the regenerative system is that this technology has particularly significant benefits in some zones of a furnace where the temperature of heating treated material is high. Furthermore, and for the same level of heat flux for both systems, the maximum temperature of the tube in the case of the regenerative system will be much lower than that in the case of recuperative system, thereby increasing the lifetime of the radiant tube.
7.9 NO\textsubscript{x} emission

Although investigation regarding the NO\textsubscript{x} emissions is not in the objectives of this thesis, the nitric oxides were observed throughout the work and measured. Figure 39 and Table 9 show the NO-emission from the three combustion configurations, single-flame conventional, single-flame HiTAC, and twin flame HiTAC parallel. NO-emissions resulted by the two HiTAC configuration are still not higher than that resulted by a conventional combustion, which is favourable in terms of NO\textsubscript{x} emission due to the low combustion air temperature (25 °C). Nevertheless, NO-emission resulted by HiTAC can be more than twice as less than any other type of combustion if the comparison considers the potential heating.

![Figure 39](image.png)

Figure 39. NO emission resulted by various combustion modes measured at the exhaust gases of a test furnace (Correlation, $\lambda = 1$).

<table>
<thead>
<tr>
<th>NO emission</th>
<th>Conventional</th>
<th>HiTAC Single-flame</th>
<th>HiTAC Twin-flame</th>
</tr>
</thead>
<tbody>
<tr>
<td>mg/Nm\textsuperscript{3}</td>
<td>83</td>
<td>78</td>
<td>83</td>
</tr>
<tr>
<td>mg/kWH</td>
<td>480</td>
<td>174</td>
<td>163</td>
</tr>
</tbody>
</table>

Conversely, from the local NO measurements and in the twin-flame configurations, it was observed that nitric oxide is rapidly generated to about 40 ppm at 350 mm downstream from the fuel gas jet (opening 6 in Figure 5). Then the NO molar fraction was levelled off downstream to this point until it exited the furnace at 55 ppm in the cases of parallel and stagger configurations, and at 75 ppm in the case of counter configuration. The results may indicate that a prompt mechanism is the dominant one since the rapid generation is in the rich fuel region in which combustion is happening between the fuel and the excess oxygen that is entrained with the combustion products into the fuel jet. Moreover, in the counter configuration, the more intense combustion and consequently the higher the fluctuation in temperature may be the reason of a higher thermal NO\textsubscript{x} generation.

On the one hand and for the HiTAC application in an indirect system, NO\textsubscript{x} measurements indicated that the regenerative system of the radiant-tube, the configuration of Figure 8g, operated at higher
efficiency compared to the recuperative system, the configuration of Figure 8f, without the cost of excess higher NO emissions. For example, the NO emission from the conventional recuperative system, calculated for $\lambda=1$, was 179 ppm and 284 ppm in tests CRS-1 and CRS-2, respectively. These emissions were quite similar to the regenerative system where NO concentrations of 167 ppm and 316 ppm were recorded for RS-1 and RS-2 tests, respectively. In fact, this relatively high NO emission result from a HiTAC-based RT burner is due to OdC criterion. According to an index made by Hasegawa et al. [12], the OdC operating condition for radiant tube application is categorized in the medium-high temperature region, 800-1000°C, and at about 12% oxygen in the combustion air. This means relatively low gas-recirculation, probably due to a small confined space around the flame. Although these values are lower that those generated by a typical HiTAC-based direct firing system, the NO emission per the effective energy from the RT is much lower in the case of a HiTAC-based regenerative system than that in the case of a conventional recuperative system.
8. Conclusions

1- Theoretical and experimental investigations throughout this work showed that the thermal and the thermodynamic efficiencies of combustion increase with a decrease in reactants’ oxygen concentration and reactants’ temperature increase.

2- Thermodynamically, the temperature increase due to combustion under OdC conditions is limited, and the irreversibility is reduced with reducing oxygen content and/or increasing the temperature of the reactants, indicating great potentials of this combustion in applications beyond those of the heating furnaces. As a conclusion, the efficiency increase of a heat engine using Oxygen-diluted Combustion would not only be due to the controlled maximum temperature by dilution that provides the possibility for an increased pressure ratio for the same maximum temperature in the combustor, but also due to the reduced irreversibility of the combustion phenomena itself. In fact, the 2nd law analysis showed that the irreversibilities can be further reduced if the oxygen-diluted combustion concept was applied using oxygen in place of air.

3- The mathematical modelling for the two major types of heat regenerators, the fixed bed and the honeycomb, were developed not only to measure and verify the thermal performance of an existing heat regenerator, but also to be a designing tool for maximized efficiency and heat recovery of the heat regenerator and thus maximize heating potential in the furnace and energy savings. The results from these models showed good agreements with measurements.

4- Local measurements have been performed in a semi-industrial test furnace equipped with HiTAC-based regenerative burning systems to analyse and characterize HiTAC flames and examine the effect of these characteristics on the heat transfer inside the furnace. The measurements of local gas composition showed, generally, a gradual disappearance of the fuel and the appearance of intermediate species hydrocarbons in relatively high concentrations and at broader regions inside the furnace in the case of HiTAC. The deep penetration of the high speed fuel and air jets into the furnace was proved by detecting the fuel downstream the burner to 1.2 m. This is evidence of a large chemical flame volume, i.e., a large reaction zone. The flame volume has been identified for single-flame HiTAC and twin-flame HiTAC configurations and was found larger than a conventional flame by 37 and 25 times, respectively. Moreover, the detection of hydrogen in high concentration and in a broad region is an indication of slow cracking of the fuel and low specific combustion intensity, and therefore low specific fuel energy release.

5- In addition to the chemistry profiles uniformity, the temperature profiles were also more uniform in the case of HiTAC configurations; no high gradients of temperatures were found. The thermodynamic limitations to the maximum temperature of the Oxygen-diluted Combustion, the low specific energy release of the fuel, and the high heat release of this large flame to its surroundings caused this uniform and relatively moderate temperature profile in the HiTAC flame, and hence suppressed thermal-NO formation. The low values of fluctuations in temperature can be a result of the non-intense presence of unburned fuel/air mixture at any measured location. In other words, reduced specific combustion intensity.

6- The above conclusion was found to be more or less valid for all HiTAC configurations having straight flame axis. However, they are also valid in the twin-flame-counter configuration, where the flame axis is curved, but to a lesser extent.

7- Local heat fluxes measurements were made to quantify the convective heat transfer coefficient in the test furnace. At a certain and typical condition, location, and configuration for heating, the convective heat transfer might be 30% of the net total heat exchange between the surface of the total heat flux meter and the hot environment in the furnace. The furnace thermal efficiency increased by 35% and 44% in the single-flame HiTAC and twin-flame HiTAC, respectively, compared to that in the conventional configuration. Nevertheless, the heating potential can be
significantly increased to more than two and three times in the case of single flame HiTAC and twin-flame HiTAC, respectively. This depends on heat recirculation performance, implying much more energy savings and consequently less pollutant emissions than the apparent energy recovery from the heat recirculation. Therefore, it is certain that this large HiTAC flame emits more thermal radiation to its surroundings than the conventional flame does, although the peak temperature of the flame is low and the temperature profile thereof is moderate-uniform. In addition, this intense heat flux was also more uniform in all HiTAC configurations than that of the conventional-air combustion configuration.

8- The HiTAC concept can be applied in the radiant tube application, to a certain extent. The temperature uniformity of the tube, caused partly by the twin burner configuration and partly by the HiTAC phenomena, resulted in increased heat flux intensity and uniformity. Furthermore, the use of oxygen-diluted combustion in a radiant tube contributes in increasing the lifespan of the tube.

9- In addition to the reduction of pollutants emissions from the above energy savings potentials, the absolute NOX-emissions (mg/Nm³) resulted by Oxygen-diluted Combustion was even lower than that generated by conventional combustion using ambient air as an oxidizer.
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