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# Analysis of Oxy-Coal Combustion through Measurements in a Pilot-Scale Entrained Flow Reactor

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Coal combustion is investigated in both air and oxy-fuel conditions in a pilot-scale entrained flow reactor able to provide high temperatures, heating rates and residence times. Measurements are carried out with different levels of complexity and are aimed at: assessing the thermal field inside the reactor; evaluating conversions of devolatilization or char combustion tests; identifying phenomena such as volatiles ignition and measuring the ignition delay time. Computational Fluid Dynamics was also used in order to provide a better understanding of the experimental evidences. Among the results, the ignition delay time was found to be larger in oxy-fuel conditions than in air, mainly because of the larger specific heat of the oxy-fuel environment. The proposed investigation may help the qualification of advanced experimental apparatus as entrained flow reactors, with the purpose to make them suitable for heterogeneous kinetics studies in oxy-fuel conditions.

## 1. Introduction

It is believed that coal will still play a fundamental role in the future for electric power production because of its low cost and abundance; hence, recently large attention is paid to oxy-fuel combustion as a $CO_2$ capture and storage (CCS) technique for reducing greenhouse gases emission from coal plants. In oxy-fuel combustion, a mixture of oxygen and recycled flue gases is used instead of air for fuel oxidation; consequently, a gas consisting of $CO_2$ and $H_2O$ is obtained, with a $CO_2$ concentration ready for sequestration. However there is lack of information on coal reactivity in oxy-fuel conditions (Wall et al., 2009).
Hence some studies in literature have been aimed at understanding solid fuel combustion in oxy-fuel conditions through the use of advanced experimental facilities able to provide data under operating conditions (i.e. temperatures and heating rates) similar to industrial ones. Conventional analysis (e.g. thermogravimetry) on lab-scale can only give a fingerprinting of the fuel, because the thermal conditions are far from those of practical applications. For instance, pyrolysis kinetics changes substantially when varying the heating rate.
Conversely, entrained flow reactors (EFRs) are particularly interesting as they allow studying solid fuel combustion at high heating rates, (of the order of $10^3$-$10^4$ K/s) and temperatures. Different analyses can be carried out on the gaseous products (speciation) and solid residues (char properties), so they are frequently used for determining solid fuel conversions in specific operating conditions. However they could potentially provide other important information (for example kinetics) but only with the aid of sophisticated experimental techniques or tedious procedures to determine the effective particle thermal history (Simone et al., 2009a; Simone et al., 2009b).
The present work describes the on-going activities at the International Flame Research Foundation, on the characterization of oxy-coal combustion through the use of a pilot-scale entrained flow reactor, able to provide high temperatures, heating rates as well as residence times. In particular some activities were aimed at investigating the ignition delay time of dense streams of particles in oxy-fuel conditions. Although particle group effects are likely to play a strong role for the flame-holding process in practical pulverised coal burners, most of investigation on oxy-coal combustion with entrained flow reactors analyze the

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behavior of a single solid fuel particle (e.g. Shaddix and Molina, 2009; Bejarano and Levendis, 2008; Zhang et al. 2010, Simone et al. 2009, Jovanovic et al., 2011; Khatami et al., 2012) and there is lack of data on the combustion of dense streams under oxy-fuel conditions.

## 2. Experimental campaigns

### 2.1 Entrained flow reactor

The entrained flow reactor is called Isothermal Plug Flow Reactor. It belongs to the International Flame Research Foundation (IFRF) and is sited in the Experimental Area of ENEL Ricerca in Livorno. A scheme of the EFR is shown in Figure 1 (Biagini et al., 2010). It allows testing solid fuels under conditions similar to industrial applications with high temperatures (1000-1600 K) and heating rates ($10^4$-$10^5$ K/s). The residence time is up to 2 seconds. The reactor inner tube is 4.5 m long with a diameter of 0.15 m. At the walls, nine modules with electric resistances keep the temperature at a set point value. Each module has several ports which are available for coal injection or for the insertion of measuring instruments. Pulverized fuel particles are transported by a carrier gas (nitrogen/air) and injected from a side through a radial into a flue gas stream, from a pre-heating combustion section, and move along the reactor. At the reactor bottom, flue gas and particles are quenched down to 500 K with nitrogen and then treated in a separation and analysis system made of two cyclones and a bag filter, before being discharged.

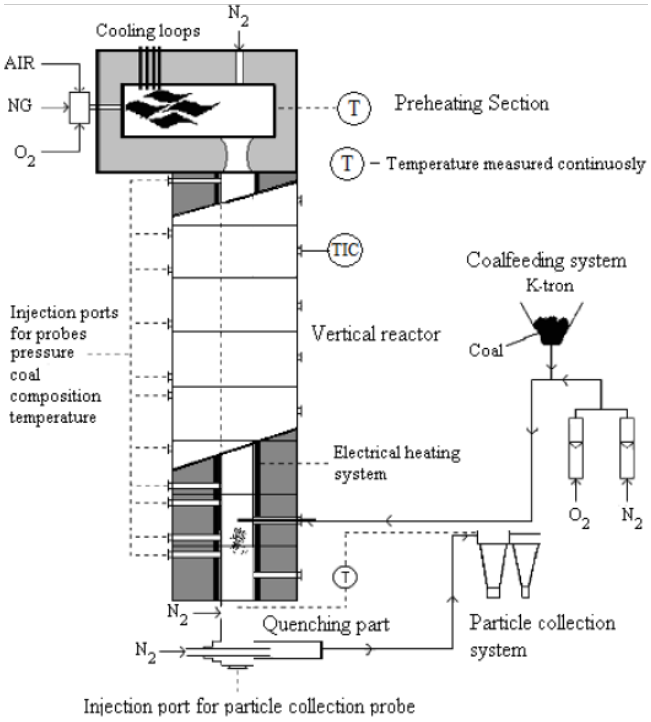


*Figure1 : (a) Scheme of the entrained flow reactor.*

### 2.2 Experimental measurements

The experimental runs regarded both devolatization and char oxidation tests. Different experimental runs were carried out, corresponding to:

STEP 1: characterization of the thermal field inside the reactor;

STEP 2: measurements of conversion from tests of devolatilization of the coal or from tests of char combustion from the output sample from the reactor;

STEP 3: measurements of the radiant energy emitted from the combustion process.

The latter was performed with ODC (Optical Diagnostics for Combustion) probes developed by ENEA (Giacomazzi et al., 2008) which were applied for the first time to coal combustion in the context of the present work. The ODC system is based on a photodiode which detects the radiant energy emitted by a combustion process in the spectral range from the UV to near IR (200 nm to 1100 nm). For gas

combustion, such energy is the result of chemiluminescence of reactants and emission/absorption from combustion product. In case of coal combustion the radiant energy may come from the gaseous combustion process (volatiles oxidation), soot as well as from burning char particles.
Four ODC probes were used and their spatial arrangement was studied in order to allow correlating different information in time and space. In particular 3 probes were inserted from the lateral ports at different distances from the feeding probe and one probe was inserted from the bottom of the reactors. The probes are not intrusive as their upper edge is placed exactly at the reactor walls.
When using the ODC probes, the feeding of the coal particles was made in a pulsed manner: a constant volume of fuel (250 mm3) was injected into the IPFR every 6 seconds in order to have pulses of groups of particles well separated from the others. The cold flow group number G was ranging from 9.7 to 40, thus indicating dense conditions (Galletti et al., 2013), At least 40 pulses were fed, for each operating condition in order to evaluate data uncertainties. The ODC signals were analyzed to evaluate the particle ignition delay time. More details about the experimental methods and signals processing from ODC technique may be found in Galletti et al. (2012) and Galletti et al. (2013).
The experimental runs were made with different coal sizes (38-90 μm and >125 μm), different concentrations of $O_2$ (from 0 to 9% by vol.), and different temperatures (from 1173 to 1573K). Details of the experimental campaign are reported on Galletti et al. (2012) and Biagini et al. (2010). The coal was a Southern African Coal; proximate and ultimate analyses are reported in Table 1.

*Table 1: Proximate and ultimate analyses of the South African Coal.*

| *Size [μm]* | *Moisture [%]* | *VM [% db]* | *FC [% db]* | *Ash [% db]* | *C [% db]* | *H [% db]* | *N [% db]* |
|---|---|---|---|---|---|---|---|
| 38-45 | 2.47 | 23.89 | 58.87 | 17.24 | 68.36 | 0.87 | 1.59 |
| 38-90 | 2.37 | 28.01 | 53.45 | 16.55 | 68.10 | 4.10 | 1.53 |
| 90-125 | 1.23 | 25.16 | 58.84 | 16.00 | 67.93 | 2.54 | 1.53 |

## 3. Numerical model

As mentioned before, experiments were complemented by Computational Fluid Dynamics (CFD) modeling.
The CFD model was based on a two-way coupled Lagrangian tracking and was developed with the commercial CFD code Ansys 13, using the fluid dynamic software Fluent.
The grid was generated with the software ICEM. Just half IPFR was modeled because of its geometric symmetry. In order to avoid creating one geometry and one mesh for each experimental run, the chosen domain referred to the run with the higher reactor length (i.e. higher distance between feeding piper and sampling probe). A grid independency study on the velocity field leaded to 900k elements.
The CFD modeling was performed with different levels of complexity in order to gain insight into specific flow features. Hence, different simulations were carried out corresponding to: single-phase flow, injection of inert particles and injection of reactive particles (coal/char).
Reynolds-averaged Navier-Stokes equations were solved using the standard and RNG κ-ε turbulence models. Different devolatilization models were applied: a Single First Order Rate (SFOR), a 2-step Kobayashi model (Kobayashi et al., 1976) and the Chemical Percolation Devolatilization (CPD) model (Fletcher et al., 1992). The Eddy Dissipation Model was used to treat the turbulence-chemistry interaction of the gaseous phase. Radiation was taken into account through the P1 radiation model and Weighted Sum of Gray Gases Model (WSGG) for the spectral properties. The particle emissivity was 0,7. The char oxidation was evaluated with the Kinetic/Diffusion Surface Reaction Rate model (Baum and Street, 1971).
A stationary solver was used to solve the equations using a second order discretization scheme and the SIMPLE algorithms for the pressure-velocity coupling. The steadiness of the solution with iteration was checked for convergence. All residuals were usually below $10^{-4}$.

## 4. Results and discussions

Measurements of the thermal field (STEP 1) inside the reactor, without the injection of coal, were run for assessing the correspondence to the nominal temperature. Figure 2a shows the comparison of predicted and experimental temperatures at different distances from the feeding probe. It can be observed that the temperature minimum is not placed in the reactor axis but it is shifted towards the sample probe side. The

reason may be imputed to the shape of the feeding probe, as can be derived from the thermal field predicted from the CFD model shown in Figure 2b.

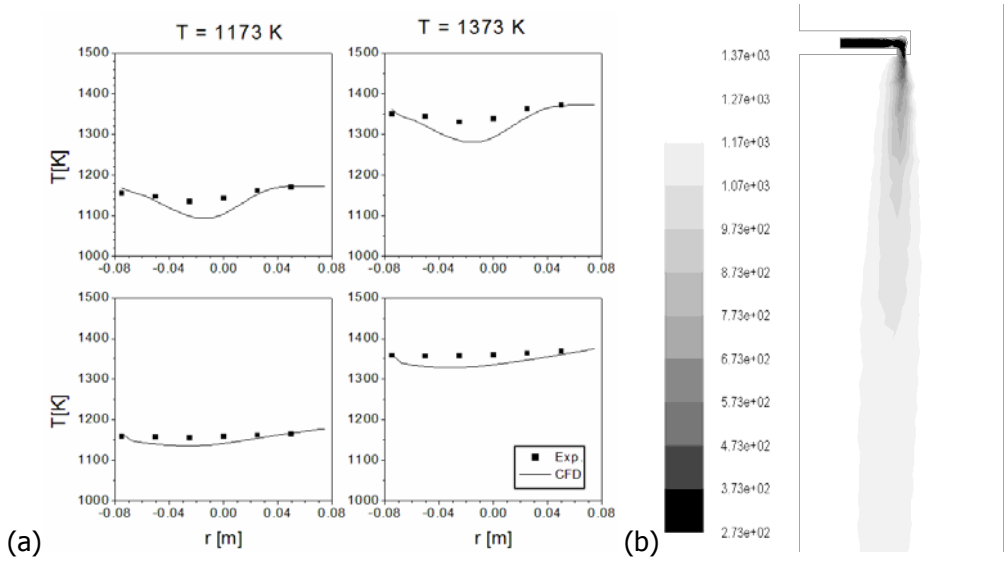


*Figura 2: (a) Radial profiles of temperature obtained at different distances from the feeding brobe (0.51 m and 1.01 m for bottom figures) and (b) Thermal field (in K) predicted with CFD for T = 1373 K.*

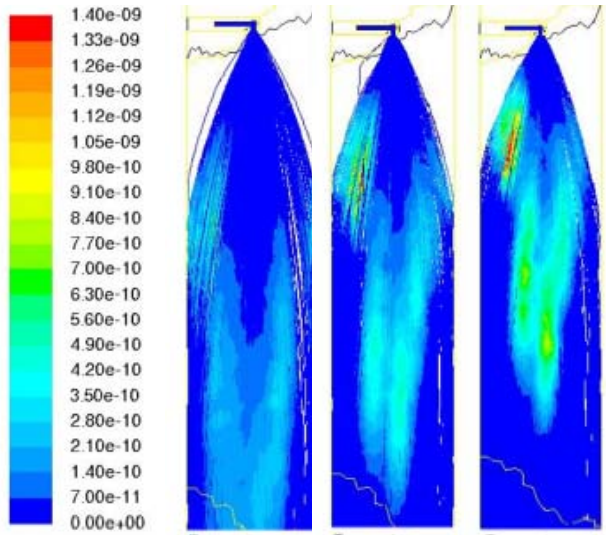


*Figura 3: Devolatilization rate for T = 1173, 1374 and 1573 K as predicted from CFD simulations using the the SFOR devolatilization model. Air combustion experiments.*

Figure 3 shows coal trajectories colored by the devolatilization rate for different nominal temperatures. It can be observed that, although only the particles near the reactor axis are likely to be sampled, many coal particles deviate from the carrier gas towards the reactor walls, resulting in an earlier heating and devolatilization.

Figure 4 shows the comparison between predicted and measured conversions (STEP 2). Mean, minimum and maximum conversions of the sampled coal particles are reported for the predictions. Significant deviation between experimental and measured conversions is observed, thus demanding for an improvement of heterogeneous kinetics. The use of a 2-step or the CPD model did not ensure any significant improvement.

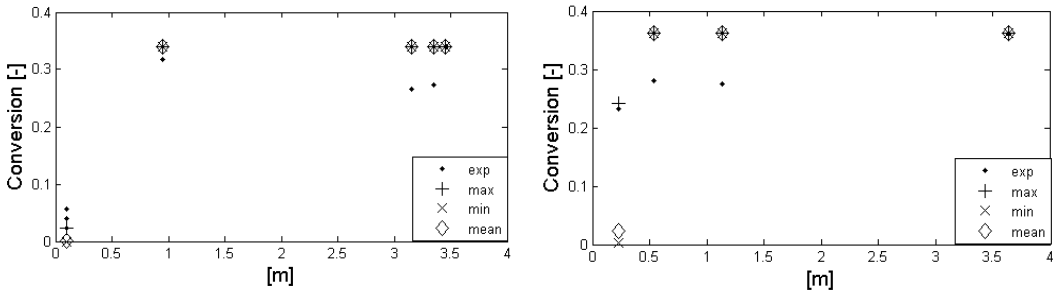


*Figure 4 – Experimental values of conversion of the particle and the maximum, minimum and mean values of those predicted with SFOR devolatilization model for reactor temperatures: (a) T=1173K (b) T=1573K. Air combustion experiments.*

Figure 5a represents the ignition delay measured with probes ODC (STEP 3) in both air and oxy-combustion conditions at the temperature T = 1373 K and with particle size of $d_p$ = 35-90 μm. The ignition delay decreased with increasing the oxygen content. Moreover, larger ignition delay times were observed in oxy-fuel conditions ($CO_2$ diluent) than in air condition ($N_2$ diluent), in agreement with the work of Shaddix and Molina (2009). This may be imputed to the higher heat capacity of $CO_2$ with respect to $N_2$.
It can be shown from the adiabatic thermal explosion theory for a one-step overall reaction, that the auto-ignition time increases linearly with the specific heat according to the following equation (Shaddix and Molina, 2009) :

$$\tau_i = \frac{c_v\left(T_0^2/T_a\right)}{q_c Y_{F,0} A\exp(-T_0^2/T_a)} \qquad (1)$$

where $c_v$ is the specific heat at constant volume, $T_0$ is the initial temperature of the fuel/oxidiser mixture, $q_c$ is the combustion heat release per mass of fuel, $Y_{F,O}$ is the initial mass fraction of the fuel and the reactivity of the fuel/oxidiser mixture is given by $k = A\exp(-T_a/T_0)$.

Figure 5b shows the ignition time to molar specific heat ratio for both air and oxy-fuel conditions. The molar specific heat has been evaluated at 1373 K for $O_2/N_2$ and $O_2/CO_2$ mixtures. It can be noticed only minor differences (of the order of the uncertainty) between air and oxy-fuel conditions, thus confirming that the main reason for higher ignition delay in $CO_2$ atmosphere is due to the higher heat capacity of $CO_2$.

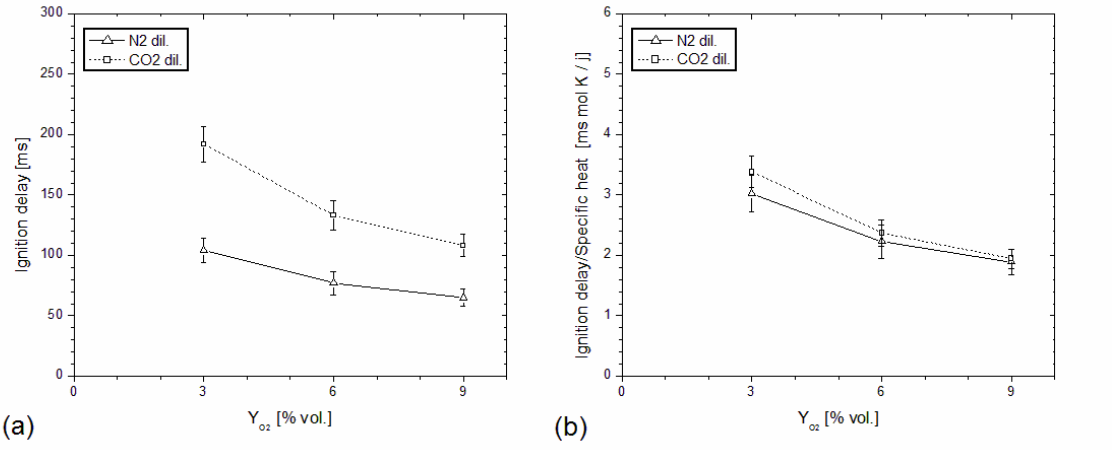


*Figure 5: (a) Ignition delay and (b) ignition delay to specific heat ratio as a function of $O_2$ fraction for different diluent gases. Reactor temperature T = 1373 K , particle diameter $d_p$ = 38-90 ν m.*

## 5. Conclusions

A pilot scale entrained flow reactor is used for studying coal pyrolysis in conditions of air combustion and oxy-combustion. Different experiments have been made in order to qualify the reactor and make it suited for conversion studies as well as ignition delay time investigation. CFD modelling of the reactor was also perform to allow a better interpretation of the experimental data as well as to provide particles thermal histories which are needed to derive kinetics.

Preliminary results indicated that the ignition delay is higher in oxy-fuel conditions than in air conditions, mainly because the higher heat capacity of $O_2/CO_2$ mixtures than $O_2/N_2$ mixtures.

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