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Characterization of solid fuels in oxy-fuel conditions

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Oxy-fuel combustion is an option for carbon capture and sequestration. This technology is still at the demonstration stage and there are several research needs that should to be satisfied to improve the reliability of this technology. One of the key issues to address is the behaviour of solid fuels switching from traditional air firing to oxy combustion. In this work a methodology to evaluate the properties of solid fuels is described. The experimental work is coupled with a CFD model of the DTR to predict the thermal histories of the coal particles, which can be used to evaluate kinetics according to a previously developed routine. Some preliminary results of firing tests at high heating rate of a Columbian coal with atmospheres at different CO_2/O_2 ratios are presented. The tests are carried out in a Drop Tube Reactor operating at a nominal temperature of 900 °C. Tests in air and nitrogen are performed for comparison. The results show that in these operating conditions carbon dioxide reduces the conversion of the coal particle, therefore a higher proportion of oxygen is required in Oxy-fuel conditions to match the conversion obtained from air firing.

1. Introduction.

The geopolitical distribution of coal reserves compared to those of oil and gas, as well as the larger supply base, indicates that coal is likely to become the main energy source for power generation in the near future. However there are environmental concerns on coal utilization especially because of its high C/H ratio which leads to large CO_2 emissions. To achieve a significant reduction, the CO_2 generated from coal utilization needs to be captured and stored (sequestered). Several CO_2 capture and sequestration (CCS) technologies have been proposed such as those based on the scrubbing of gases from pulverized coal plants (post-combustion), integrated gasification combined cycles with air separation units (pre-combustion) or oxy-fuel combustion (Wall, 2007). In the latter technology coal combustion occurs in oxygen rather than air, and the oxygen is diluted with an external recycle of flue gas (see Figure 1), so that to reduce the

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combustion temperature and add gas for the following heat transfer operations. This is an effective solution to produce a CO_2 rich stream, ready for capture and sequestration.

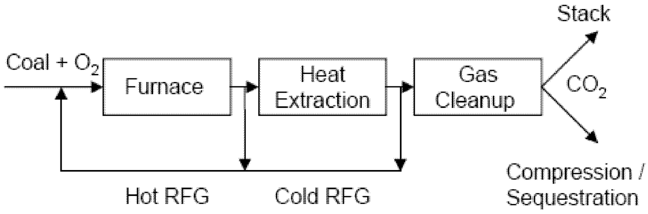

Figure 1-General flow-sheet for oxy-fuel combustion.

It is recognized that, compared to air firing conditions, the high content of carbon dioxide in oxy-fuel conditions may influence the combustion of coal in several manners (Buhre et al., 2005; IFRF 31st TOTEM):

- a higher proportion of oxygen is required to match the same adiabatic flame temperature;
- higher gas density and emissivity ;
- higher content of corrosive species;
- potential reduction of NOx formation;
- potential increase in soot formation;

However oxy-fuel combustion is still at the demonstration stage and there are many research needs (IFRF 31st TOTEM) that should be addressed:

- investigation on high temperature NOx reduction mechanisms;
- development of soot formation models and soot radiation properties;
- impact of coal devolatilization on char properties ;
- char oxidation mechanisms and kinetics;
- co-firing with biomass;
- ash behaviour, fouling and corrosion;
- optimized recycled flue gas ratio and oxygen content

Among others institutions the International Flame Research Foundation (IFRF) is dealing with the above mentioned research needs pursuing and developing several activities according to a problem decomposition technique (see Figure 2). All the levels of the pyramid contribute to fully characterise a complete semi-industrial system: the Fo.Sper furnace at the ENEL research area of Livorno retrofitted to Oxy-Fuel conditions. The investigation of the behaviour of solid fuels in oxy-fuel atmospheres is one of the main issues of this approach and it is fundamental to provide reliable data for modelling and designing oxy-fuel systems as well as retrofitting old ones. To this purpose the IFRF is planning an experimental campaign with an Isothermal Plug Flow reactor. This preliminary work describes a methodology to characterize the behaviour of solid fuels in different gas atmospheres. Tests focusing on the first devolatilization at high heating rate of a Columbian coal under different CO_2/O_2 atmospheres are carried out in a lab-scale Drop Tube Reactor (nominal temperature of 900 °C). Tests in air and nitrogen are performed for comparison.

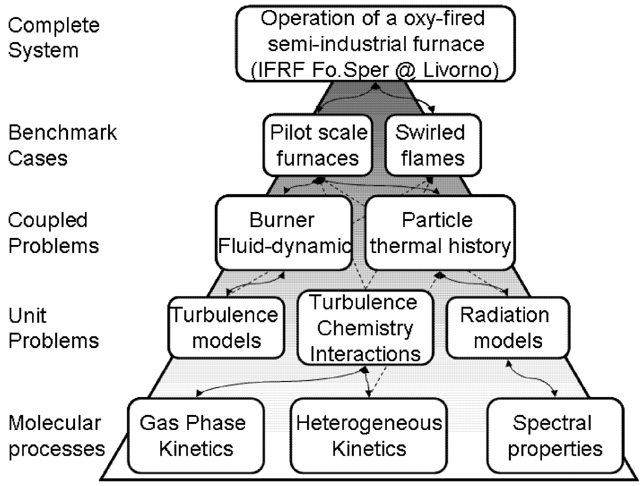

Figure 2-Hyerarchic approach for problem decomposition of Oxy-Fuel combustion.

To obtain information about the coal conversion kinetics, the particle thermal histories inside the DTR are to be known. These are derived from a DTR model made with Computational Fluid Dynamic (CFD) which can also provide a characterization of the operating conditions in the DTR.

2. Experimental system

The DTR is an Inconel 600 tube inserted in a vertical electric heater formed of three independent resistances (Figure 3). The feeding system is positioned at the top of the DTR. A gas stream (4 Nl/min) is fed coaxially and flows downward entraining the solid particles, which are continually forced by an electric motor through a lateral tube.

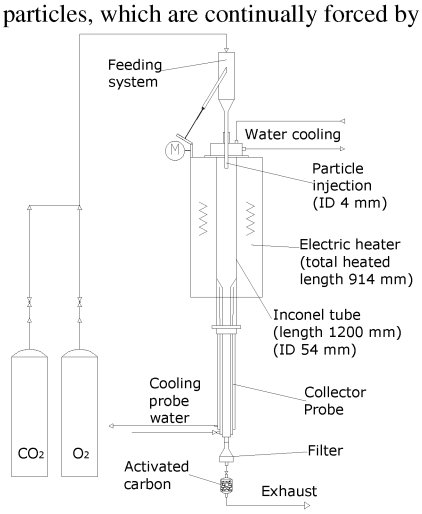

Figure 3-Scheme of the experimental system.

The collector probe cools down the gas and stops the reactions of the solid. This latter is collected on a glass fibre filter for off-line analysis. A Columbian coal (VM 38.4, FC 53.7, ash 7.9%wt dry basis) milled and sieved to the dimensional range dp = 63 – 80 µm is used in the tests. The firing tests are carried out at nominal temperatures of 800 °C, 900 °C and 900 °C on each resistance, respectively. The reactor length is kept constant at the maximum value (914 mm). Five gas atmospheres were tested 70%CO_2-30% O_2, 80%CO_2-20% O_2, 100% CO_2, Air and 100% N_2 for comparison. The solid residues produced from tests in the DTR were subsequently oxidized with air to obtain the conversion according to the “ash tracer” method.

3. Experimental results

Figure 3 reports the conversion of the coal due to the thermal treatment in the DTR under different atmospheres. The lowest conversion value (16%) is obtained in the test with 100% CO_2. When oxygen is added to the gas stream a higher value is achieved and the higher the oxygen concentration the higher the conversion value. All the conversion values achieved in the DTR tests are lower than the content of organic material in the coal reported in the proximate analysis. Therefore the short residence time of the particles in the DTR does not allow a complete conversion of the organic material (at the nominal temperature of 900 °C) and the coal is subjected to a first devolatilization treatment. In addition the effect of nitrogen and carbon dioxide are compared performing a pyrolysis test in pure nitrogen and a firing test in air. The conversion value obtained with pure nitrogen is slightly higher than the value achieved in the test with pure carbon dioxide (see Figure 3). As a matter of fact the standard deviation of the conversion is quite high for both tests, so the results can be considered comparable within the experimental errors. The test with 80% CO_2 and 20% O_2 simulates the substitution of nitrogen in air with an equal volume of carbon dioxide. The conversion value obtained in this test (20.6%) is lower than the one obtained in the test with air (23.2%). Hence, carbon dioxide in these operating conditions leads to a reduction of the conversion and the gasification reactions are likely to be negligible.

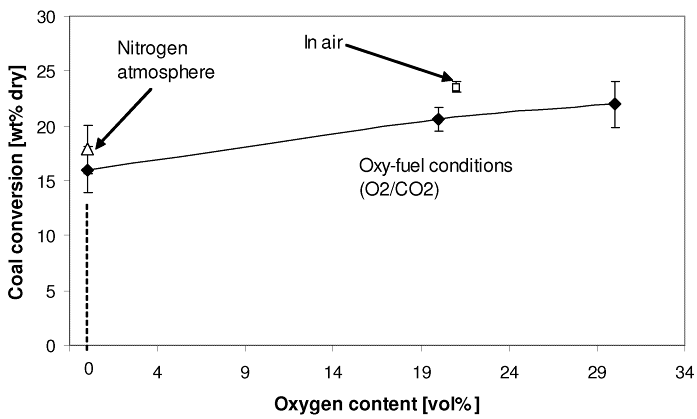

Figure 3-Coal conversions achieved from DTR tests in different gas atmospheres.

The above mentioned results are in agreement with findings of other authors (Borrego and Alvarez, 2007) which suggest that carbon dioxide could be involved in cross-linking at the particle surface. The conversion value obtained in air is approached in the test with 70% CO_2 and 30% O_2. This oxygen proportion can be used in practical systems to attain an adiabatic flame temperature similar to air firing (Buhre et al., 2005). Therefore to match coal conversion in the DTR achieved during air firing tests, the oxygen content in the CO_2-O_2 test must be increased.

4. CFD aided experiments

Firing tests in different gas atmosphere can give important information about how these will affect the fuel behaviour in the system. However in order to calculate reaction kinetics the thermal history of the fuel in the DTR is to be known. To this purpose the experimental work can be aided with a CFD model of the DTR. The model was previously developed to evaluate the particle thermal history of biomass fuels during pyrolysis in nitrogen to calculate high heating rate devolatilization kinetics [Simone et al., 2008a]. In this case the model has been applied to coal firing both in air and oxy-fuel atmospheres. The coal reactions (devolatilization, char and gas oxidation) were represented with default sub-models and data related to air firing available in the code Figure 4a reports the thermal history predicted by the CFD model of a coal particle in the DTR under two gas atmospheres 70% CO_2 -30% O_2 (named Oxy) and Air. These preliminary results show that the particle heating rate ($2*10^{4}$ °C/s) seems not be affected by the gas atmosphere at the nominal temperature of 900 °C. Conversely the high concentration of carbon dioxide leads to higher residence time (2.6s vs 2.2s) and slower cooling rate than in Air due to the larger gas density and heat capacity in Oxy conditions.

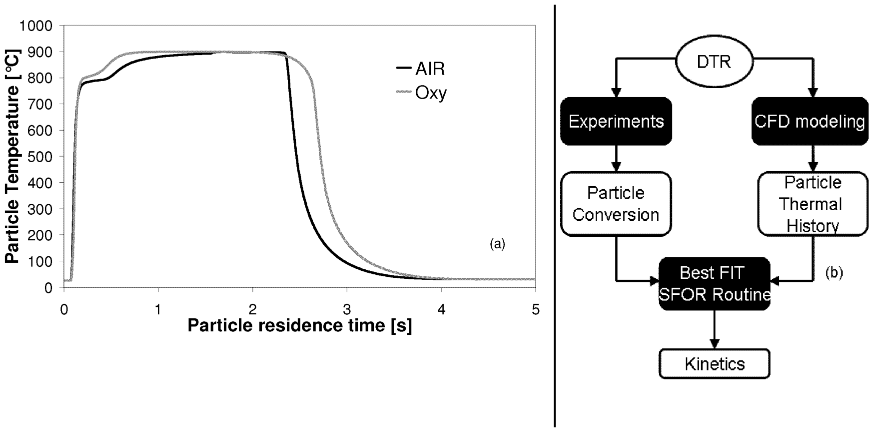

Figure 4-(a) Thermal histories of a coal particle in the DTR under two gas atmospheres (70% CO_2- 30% O_2 and Air) and nominal temperature of 900 °C , (b) methodology for calculating kinetics from experiments in the Drop Tube Reactor .

Once the particle conversions are determined from experimental tests and the particle thermal history is evaluated from the CFD model, it is possible to calculate kinetic parameters for a single first order reaction sub-model according to a best fit routine (see Figure 4b).The reliability of the kinetics can be improved by refining the assumptions of the CFD model or selecting a different reaction sub-model (Simone et al., 2008b).

5. Conclusions

A preliminary work of characterization of solid fuels in Oxy-fuel conditions has been described. The effect of carbon dioxide in Oxy-fuel combustion of a Columbian coal in a DTR has been investigated by performing tests with atmospheres at different CO_2/O_2 ratios. Tests in air and nitrogen have been carried out for comparison. The experimental results show that carbon dioxide may reduce the conversion of the coal particle in the investigated operating conditions. Therefore a higher proportion of oxygen is required in Oxy-fuel conditions to match the conversion obtained from air firing. The experimental work is coupled with a modelling activity to determine the effective thermal history of the particles in the DTR. The model of the DTR has been developed with a CFD code. Simulations of the tests in the DTR show that the particle heating rate ($2*10^4$ °C/s) is not very affected by the gas atmosphere but some differences exist in the time to reach the maximum temperature and cooling rate. Future experimental works will aim at applying the procedure to a wider range of operating conditions, gas atmospheres and fuels as well as characterizing the solid residues with previously assessed methodology (Biagini et al., 2009). The CFD model will be used also to evaluate kinetic parameters in Oxy-fuel conditions from DTR tests according to a procedure previously developed for pyrolysis (Simone et al., 2008a).

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