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Natural gas oxy-combustion with flue gas recycling for CO_2 capture

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The purpose of this work is to study and compare combustion of natural gas in air and oxy-combustion with flue gas recycling in order to optimize this technology. This paper presents several experiments of natural gas combustion using air (or enriched air) or a mixture of pure oxygen mixed to recycled flue gas ($O_2/CO_2/H_2O$) as oxidizer. The trials were conducted on a 300 kW_{th} pilot-scale facility, composed of a burner, a combustion chamber, a flue gas cooler and a flue gas recycle fan. Flue gas composition, O_2 concentration in the feeding gas and the flame temperature were monitored.
This study aims at analyzing the influence of the nature of the oxidizer, O_2 concentration in the feed gas, oxygen excess and recycling ratio on flame temperature profile, heat transfer, flue gas composition and NO_x emissions.
The results obtained showed that the flame temperature with 21% O_2 concentration in the feed gas is lower for oxy-combustion than combustion in air. However, the recycling ratio can be adjusted to control the flame temperature and an O_2 concentration of 24% in the feed gas is necessary to maintain a temperature profile similar to that observed for combustion in air. It has been observed also that oxy-combustion with flue gas recycling improves the heat transfer. Finally, the results showed also that the NO_x emissions are highly reduced because of the absence of N_2 in the feed gas.

1. Introduction

During the last decades, human activities have increased the level of greenhouse gases in the atmosphere and it is well known that carbon dioxide plays a dominating role in global warming and climate change. Carbon Capture and Storage (CCS) technologies provide a rapid way to cut CO_2 emissions by capturing it from large point sources such as fossil fuel power plants. Currently, several options are available to reach this goal.
Conventional heat production units use air for combustion in which the nitrogen from the air dilutes the CO_2 concentration in the flue gas. The capture of CO_2 from such dilute mixtures using amine stripping is relatively expensive according to Nsakala et al. (2001) and Singh et al. (2003). During oxy-fuel combustion, a combination of pure oxygen and recycled flue gas (RFG) is used for fuel combustion and, thus, a flue gas mainly composed of CO_2 and H_2O is produced. According to Wall et al. (2005), the

RFG is used to control flame temperature and make up the volume of the missing nitrogen to ensure there is enough gas to carry the heat through the boiler. According to Croiset et al. (2001), Liu et al. (2003) and Châtel-Pélage et al. (2003), this technology offers other benefits, including a potential decrease in NOx emissions and lower net combustion gas volume at higher oxygen feed concentration.
The oxy-fuel combustion contributes also to a higher boiler thermal efficiency with lower sensible heat in flue gas and offers the potential to lower oxy-fuel plant investment by reducing the scale of flue gas cleaning equipment (Marin et al., 2003).
This paper describes a series of experiments conducted on the oxy-combustion of natural gas with flue gas recycling.

2. Experimental Set-up

The pilot-scale facility of natural gas oxy-combustion, used for the trials, is shown in Figure 1. It is composed of a natural gas burner, a combustion chamber, a flue gas cooler and a flue gas recycle fan. The burner has a capacity of 300 kW_{th} and is capable of firing natural gas in air, O_2 enriched air and mixtures of O_2 and recycled flue gas (RFG). The combustion takes place in the first horizontal part of the combustion chamber which has an inside diameter of 600 mm and an overall length of 1900 mm. The second vertical part of the combustion chamber has an inside diameter of 1180 mm and an overall height of 2100 mm. Both of them are cylindrical and refractory lined. Then, the product gas passes through the flue gas cooler where the outlet temperature is controlled by using 1 to 6 cooling fans of a water recirculating system. The flue gas leaves the flue gas cooler at approximately 200 °C and a part is recycled and mixed with pure oxygen for combustion of the fuel. Pure oxygen comes from a gas cylinder rack with a full capacity of 190 Nm^3.

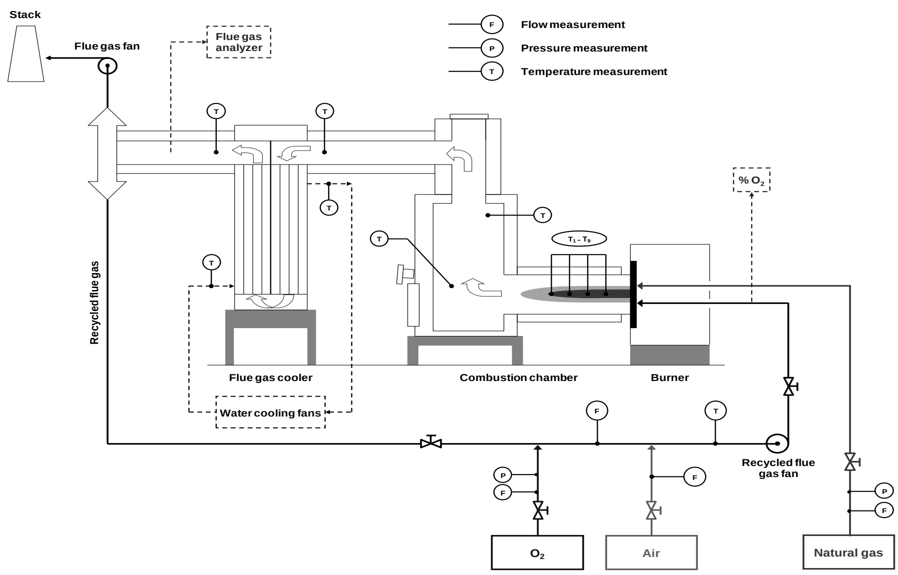

Figure 1: Pilot-scale facility of natural gas oxy-combustion

Four flow meters allow volumetric flow rate measurements of natural gas, air, pure oxygen and RFG in order to control the recycling ratio and the oxidizer composition. Thermocouples along the horizontal part of the combustion chamber permitted in-flame measurements of temperature. Four thermocouples measure the centerline temperatures whereas 5 others are shifted a distance of 100 mm. In total, 9 thermocouples measure the temperature profile in the horizontal part of the combustion chamber, 2 others are located in the vertical part of the combustion chamber, 4 give the inlet and outlet temperatures of the flue gas cooler and another one measures the oxidizer temperature. Flue gas composition (O_2, N_2, CO_2, CO, CH_4, NOx) and O_2 concentration in the feed gas (oxidizer) are also constantly analyzed.

Natural gas with a concentration in methane of 96.2% (mol. %) is used for the trials. In this paper, the term ‘oxidizer’ is used to designate alternately air, O_2 enriched air or mixtures of O_2 and RFG. The ‘excess O_2’ refers to the O_2 mole fraction (mol. % on a wet basis) in the flue gas. The ‘O_2 enrichment’ of the feed gas refers to the O_2 mole fraction (mol. % on a wet basis) in the oxidizer and may be defined as the ratio between the volume flow rate of oxygen and the total volume flow rate of the oxidizer.

Each test is conducted with a power input of 300 kW_{th} and the oxidizer is fed at approximately 100 °C. For combustion in air, the excess O_2 (O_2 mole fraction in the flue gas) is set to 2%, 4% and 6%. For combustion in O_2 enriched air, a fixed excess O_2 of 4% is used and the O_2 enrichment of the oxidizer is set alternately to 21%, 22%, 23%, 24% and 25%. For oxy-combustion trials, the excess O_2 is set to 2%, 4% or 6% and the O_2 mole fraction in the oxidizer is adjusted alternately to 19%, 21%, 24% and 26%.

3. Results and Discussion

This study aims at analyzing the influence of excess oxygen and O_2 concentration in the oxidizer on flue gas composition, flame temperature profile, heat transfer and NOx emissions. The results are compared with conventional combustion in air.

3.1 Temperature profile

Figure 2 shows the centerline flame temperature profiles measured for combustion in air. It can be observed that the average temperature is smaller when the excess O_2 increases because of the inert gases dilution effect (mainly due to N_2). Indeed, for a higher air flow rate, the combustion energy is released in a larger inert gas volume and, thus, the temperature measured decreases.

Figures 3, 4 and 5 show the centerline flame temperature profiles measured during oxy-combustion trials. It can be observed again that the average temperature is smaller when the excess O_2 increase because of the inert gases dilution effect (mainly due to CO_2 and H_2O in this case). For a given O_2 concentration in the flue gas (4%), the pure oxygen flow rate is fixed and the O_2 concentration in the oxidizer is controlled by adjusting the RFG flow rate. For a higher RFG flow rate, the volume of inert gases (mainly CO_2 and H_2O) is larger and, thus, the temperature measured decreases.

To compare oxy-fuel combustion with conventional combustion, the baseline case chosen corresponds to combustion in air with an O_2 concentration in the flue gas of 4%. From these results, it can be observed that the temperature profile in the combustor with 21% O_2 concentration in the oxidizer is lower for oxy-combustion than combustion in air.

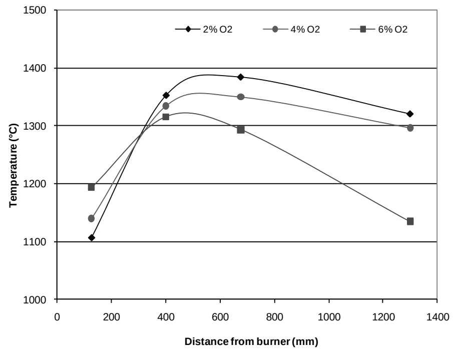

Figure 2: Temperature profiles for combustion in air (21% O_2 in air)

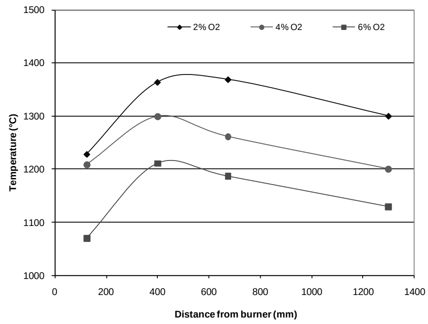

Figure 3: Temperature profiles for oxy-combustion (19% O_2 in oxidizer)

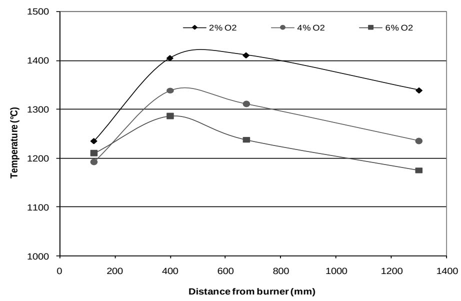

Figure 4: Temperature profiles for oxy-fuel combustion (21% O_2)

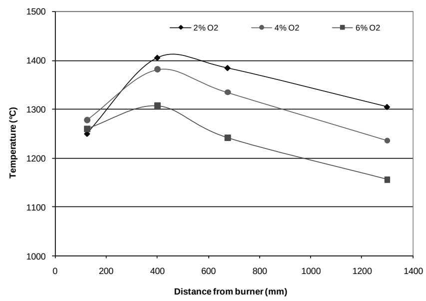

Figure 5: Temperature profiles for oxy-fuel combustion (24% O_2 in oxidizer)

Oxy-fuel combustion generates a flue gas with high proportions of CO_2 and H_2O, which have high heat capacities compared to N_2. This results in smaller average temperature in the combustion chamber. To attain a similar flame temperature to that observed for conventional combustion, the O_2 concentration in the oxidizer has to be higher than 21%. An O_2 concentration of 24% in the feed gas is found to produce the closest conditions compared to air firing.

3.2 Heat transfer

Thermal transfer in the flue gas cooler is evaluated by calculating the heat exchanged (kJ) with the water cooling system for each Nm^3 of flue gas passing through it (kJ/Nm^3). Figure 6 shows the results obtained in the range of 21−25% of O_2 in the feed gas for combustion in air and enriched air and in the range of 19−26% of O_2 for oxy-fuel combustion. An O_2 concentration of 4% in the flue gas is used in each trial. From this figure, it can be observed that the heat transfer is higher for oxy-fuel combustion (≈600 kJ/Nm^3) compared to combustion in air (≈500 kJ/Nm^3). This is due to the higher concentrations of CO_2 and H_2O, which are the gases that radiate in a flame. The high proportions of CO_2 and H_2O in the flue gas result in higher flue gas emissivity and, thus, a better radiative heat transfer in the case of oxy-combustion compared to air firing.

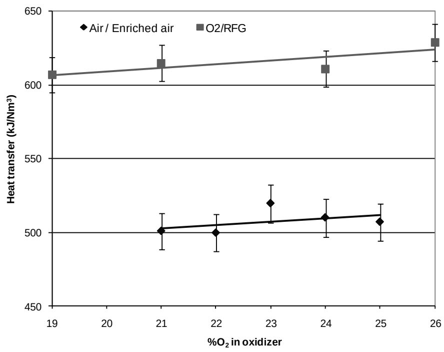

Figure 6: Heat transfer for air/enriched air and O_2/RFG combustion

3.3 NO_x emissions

Figure 7 shows the NO_x concentrations measured in the flue gas in the range of 21−25% of O_2 in the feed gas for combustion in air and enriched air and in the range of 19−26% of O_2 for oxy-fuel combustion. An O_2 concentration of 4% in the flue gas is used in each trial. In both cases, it can be seen that the NO_x concentration increases when the O_2 concentration in the oxidizer is higher. For larger O_2 concentration, the average temperature measured is higher, which increases NO_x production. It can also be observed that NO_x concentrations are smaller for oxy-fuel combustion compared to combustion in air. This is mainly due to the absence of N_2 in the feed gas and to lower temperatures in the case of oxy-combustion (for the same O_2 concentration in oxidizer). However, the NOx concentration is higher for oxy-combustion with 24% of O_2 in oxidizer compared to air firing (21% O_2). In fact, an air leakage maintains a relatively high N_2 concentration in the flue gas (approximately 10% on a wet basis). Thus, a higher NO_x reduction can be expected. Moreover, oxy-fuel combustion highly reduces the volume of flue gas after recycling. The expression of NO_x emissions in terms of concentration is not really suitable because this flue gas volume has to be taken into account. Thus, it can be said that oxy-fuel combustion reduces NO_x production rate.

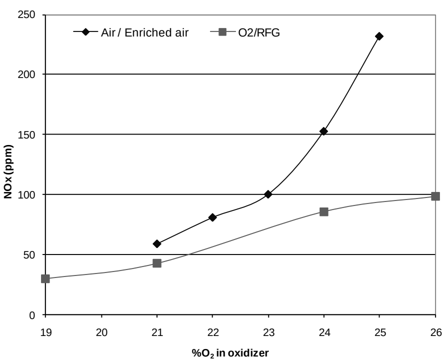

Figure 7: NO_x concentration (ppm) for air/enriched air and O_2/RFG combustion

4. Conclusions

Natural gas combustion using both air (enriched air) and pure oxygen mixed with RFG has been successfully applied in a 300 kW_{th} pilot-plant. The results showed that the natural gas oxy-combustion with flue gas recycling offers excellent retrofitting potential for conventional natural gas combustion system. Flame characteristics with oxy-combustion can be adjusted to maintain conditions similar to those observed for conventional combustion. Oxy-combustion with flue gas recycling improves the heat transfer, because of the composition of the flue gas (mainly CO_2 and H_2O), which have higher absorption coefficients compared to N_2, improving the heat transfer by radiation. With enriched air combustion, very high level of NO_x formation was achieved resulting from high flame temperature. Using oxy-combustion with no air leakage, NO_x formation could be completely suppressed due to the absence of N_2 in the feed gas.

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