



CHARACTERISTICS OF OXY-FUEL COMBUSTION USING FLUE GAS RECIRCULATION

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ملخص البحث

يتم التركيز بشكل كبير على مشاكل طبقة الأوزون في طبقات الجو العليا وتراكم ثاني أكسيد الكربون والتي تغير في طبيعة المناخ. في هذه الدراسة يتم تدوير جزء من العادم وخلطه بالأكسجين النقي لعمل خليط بديل عن الهواء في عملية الاحتراق وذلك لتخفيض انبعاثات أكاسيد النيتروجين وكذلك سهولة فصل ثاني أكسيد الكربون وتخزينه. يتم تدوير العادم خارجيا في عملية احتراق غير سابقة الخلط باستخدام الكيروسين كوقود سائل لدراسة تأثيره على ثبات اللهب وانبعثاته. مع النسب المختلفة من العادم المدار يسلك اللهب بشكل عام نفس سلوك اللهب عند استخدام الهواء الجوي. يزيد تركيز أكاسيد النيتروجين مع زيادة نسبة التدوير حتى نسبة معينة بعدها يبدأ في النقصان. فمثلا عند نسبة تكافؤ حوالي 0.8 ونسبة تدوير 40% تبلغ قيمة أكاسيد النيتروجين نفس قيمتها في الحريق التقليدي. كما تنخفض قيمة أكاسيد النيتروجين المنطلقة للجو الخارجي نتيجة لنقصان كتلة العادم الكلية المنبعثة للجو.

ABSTRACT

Attention has been focused on the breakdown of the protective layer of stratospheric ozone, and the accumulation of CO₂ that significantly change climate patterns. In the present study a mixture of flue gas recirculation (FGR) and oxygen was used as oxidizer instead of air for CO₂ capture and storage and NO_x reduction. The external FGR was employed in non-premixed combustion using kerosene as fuel to investigate its effect on flame appearance and emissions. Flame exhibits the same general appearance with different FGR% and have the main characteristics of air combustion. NO concentration increases with FGR until certain value after which goes into decrease. For instant, at equivalence ratio 0.8 the NO has the same value of reference condition at about 40% FGR. The mass of emitted NO to atmosphere were reduced as a result of exhausted mass reduction and hence the peak value of NO was shifted to lower FGR%.

Keywords: Flue gas recirculation; liquid fuel; kerosene combustion; CO₂ capture and storage; NO_x reduction

INTRODUCTION

Flue gases from fossil fuel-fired combustion facilities are major contributors to atmospheric pollution. These flue gases have high concentrations of the carbon dioxide (CO₂) beside to some concerned species, such as oxides of sulfur, oxides of nitrogen and fine dust. All these compounds are actually unwanted by-products from a power plant or a waste incinerator. So-called greenhouse gases (GHGs), most importantly CO₂ and nitrous oxide connect to measurable climate change effects such as global warming, sea-water temperature rise and more frequent extreme weather situations [1].

Direct inhalation of NO_x in high concentrations or for long periods of time can result in severe and possibly long-term damage to the lungs. Acidic deposition produced from nitrogen oxides causes damage to soils and plants, corrosion and erosion of stonework and acidification of surface waters [2].

Since fossil fuel firing will be the major heat and power source for many years to come, CO₂ capture and storage (CCS) from flue gases cannot be avoided, if such significant emission reductions are enforced. After purification, the resulting CO₂ stream can be compressed for transport and stored in deep ocean, depleted oil/gas fields, un-mineable coal beds and deep saline reservoirs. CO₂ capture through combustion processes is based upon three main routes [3, 4].

Post-combustion: achieves CO₂ separation directly from the flue gas constituents through membranes and amine scrubbing.

Pre-combustion: consists in converting fuel into a synthesis gas, composed of carbon monoxide (CO) and hydrogen (H₂). CO can then react with water to form H₂ and CO₂.

Oxy-combustion: consists in using an oxidizer mixture mainly composed of oxygen and carbon dioxide, instead of air.

The oxy-combustion with flue gas recirculation represented an ultra-clean combustion process, leading to the reduction in unburnt fuel, in addition to the lowered emissions and increased carbon dioxide concentration. The flexibility of the process, in terms of optimizing the amounts of oxidant in the various parts of the combustion process is considered to be an important feature of the technology [5].

On a large scale, for all capture systems it was found that SO_x, NO_x and PM emissions are expected to be reduced or remain equal per unit of primary energy input compared to power plants without CO₂ capture. NO_x emissions were 15% higher in the post-combustion and lower when oxy-fuel combustion (-16%) or pre-combustion (-20%) were implemented and the implementation of the oxy-fuel technology showed the lowest PM emissions [6].

Another concept, often associated to oxy-combustion, is the chemical-looping combustion (CLC) which consists of two combined loops. Reactive solid particles circulate in the first loop and separate oxygen from the incoming air through oxidation reactions. They are then transferred to the second loop where they are reduced by the fuel through an exothermic chemical process. Reduced solid particles are then sent back to the first loop to get oxidized in air. The fuel could be oxidized to about 87% and up to 88% of all carbon leaving the fuel reactor was in the form of CO₂ [7, 8].

The main advantages of oxy-combustion using FGR compared to other CCS and NO_x technologies are; wide range of flame temperature by varying rate of FGR, decreased flue gas volume compared to air-fired plants, much higher CO₂ fraction in the flue gas than in air-fired plants and great potential to reduce emissions of pollutants.

To minimize the implications on the boiler geometry and provide a similar heat transfer to the boiler furnace walls as in the case of air firing, the recirculation ratio was found to be 67.9%, which is lower than the recirculation ratio (69.3%) leading to a similar adiabatic flame temperature, and the NO formation emitted from the system were 60% lower than in the case of air firing [9].

Y. Tan *et al.* conducted a series of experiments using natural gas and different coals with air and with mixture of oxygen and FGR to facilitate CO₂ capture. The combustion with large volumes of CO₂-rich recycled flue gas offered excellent retrofitting potential for conventional combustion systems and NO_x formation was completely suppressed due to absence of nitrogen in the feed gas. The in-furnace heat transfer and temperature profiles can be made to match those for conventional combustion by adjusting O₂ concentration and flue gas recycle ratio [10, 11].

Kim *et al.* have investigated NO_x emission characteristics and flame stability of 0.03 MW and 0.2 MW oxy-fuel combustors for varying CO₂ ratio, FGR ratio, and oxygen flow rate ratio. Five different nozzle arrangement and configurations were designed to verify the feasibility of FGR technology. The oxy-fuel combustor with two aligned separate oxidizer nozzles yielded the most stable flames in a wide range of CO₂ ratios, the flame length decreased as the fuel or the oxidized velocity increased because of the increased turbulent intensity, and NO level was substantially decreased with increasing the oxidizer velocity. The reduction ratio of NO emission was approximately 85% – 93% at 40% FGR [12 – 15].

K. Andersson and F. Johnsson carried out flame radiation characteristic and burnout behavior with different recycled feed gas mixture volumetric concentrations of O₂, Oxygen/Fuel (OF) 21 @ 21% O₂ -79 % CO₂ and OF 27 @ 27% O₂-73% CO₂. The flame emissivity for both cases differs from air-fired conditions and OF 27 case exhibited similar overall combustion behavior as the air-fired reference case in terms of gas concentration and temperature profiles. The flame radiation intensity increased with up to 30% for the OF 27 case compared to air-fired conditions [16].

Andersson *et al.* have measured the radiative heat transfer and emissions in oxy-fuel flames with varying flue gas recycle rate. The temperature and the radiation intensity of the flame, increased with decreasing degree of recycle or increasing O₂ concentration in the recycled flue-gas. The reverse Zeldovich mechanism was shown to significantly reduce NO at high temperature. The amount of NO emitted during oxy-fuel operation was reduced by 70 – 75% of the emission at air-firing. [17, 18].

EXPERIMENTAL SETUP

The study was performed in a 20 kW laboratory furnace. The experimental system was designed so that air, liquid fuel, pure oxygen and FGR could be introduced, controlled and metered. It comprises a

horizontal furnace fired by laboratory designed burner equipped with facilities for flue gas recirculation. The combustion chamber is cylindrical and consists of 150 mm inner diameter pipe with 800 mm length. The combustion chamber was surrounded by a cooling jacket of 200 mm inner diameter pipe. Cooling water was delivered from the water supply network. The flow rate of cooling water was measured and controlled with a calibrated rotameter and globe valve, and its temperature was measured with a type K thermocouple and indicator. The cooling water flow rate was kept constant in each case.

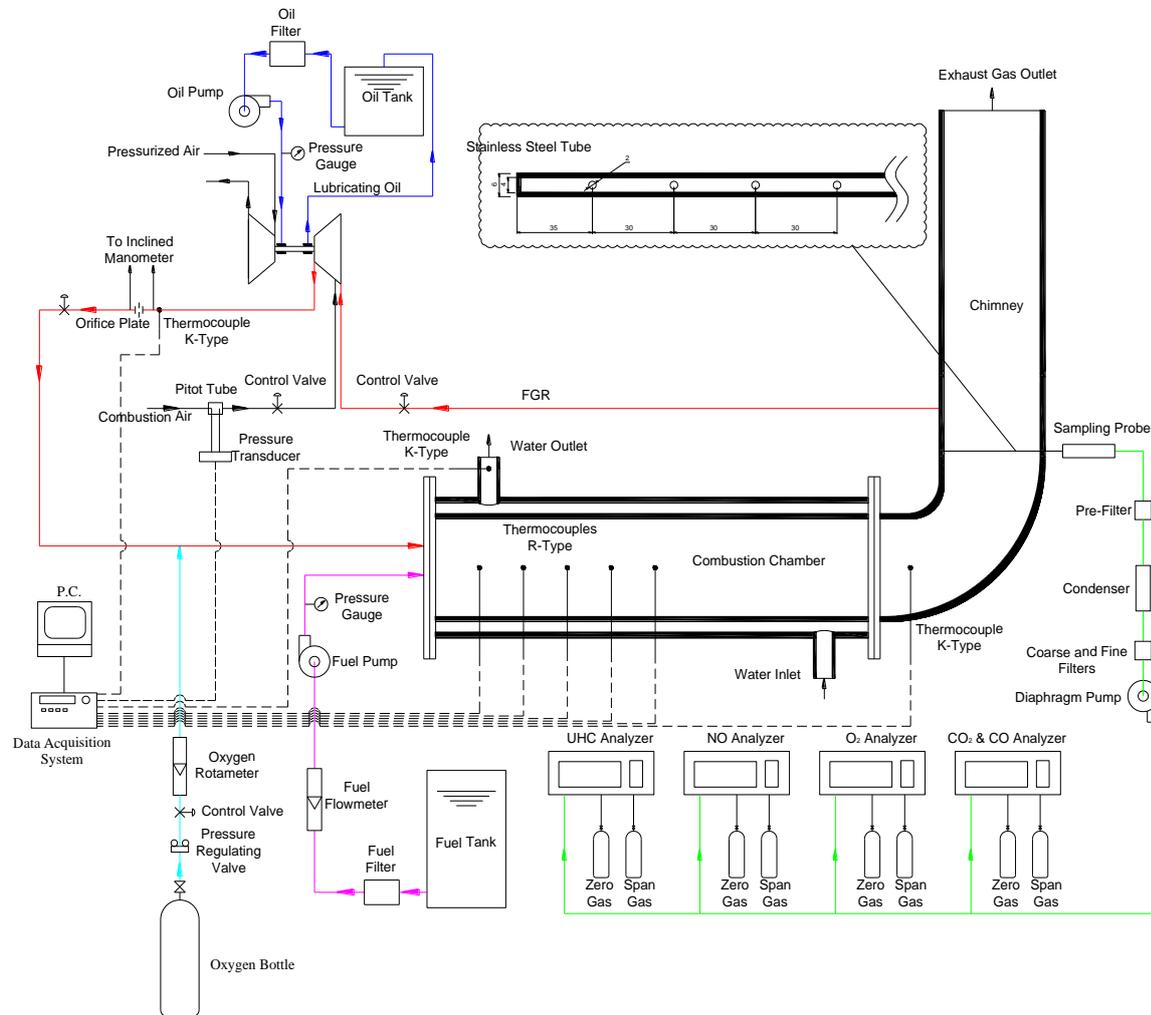


Fig. 1. Schematic diagram of the experimental apparatus.

A schematic diagram of the experimental setup is shown in Fig. 1. The kerosene was used as a fuel and atomized into a fine spray by a fuel pump and a spray nozzle. This spray was ignited by an electric spark stimulated by two solid electrodes under high voltage (10000 V). The liquid fuel (kerosene) was measured by a calibrated rotameter. The fuel flow rate was controlled via a pressure bypass valve which is a part of the fuel pump. A turbocharger was used to withdraw both combustion air and recycled flue gases. The turbocharger is reversed in operation i.e. the turbine side is become the suction side while the compressor side is the prime mover by the use of pressurized air.

The material of the turbine blades (Incoloy alloy) is high corrosion and heat resistance, therefore it is suitable to be used in flue gas applications. The speed of rotation was controlled by adjusting the flow rate of pressurized air, which was controlled by a control valve and a bypass. Both of combustion air and recycled flue gases paths were equipped with control valves to adjust flow rates. The combustion air flow rate was measured by a Pitot tube and the differential pressure was measured with a pressure transducer of range 25 mm H₂O. The transducer output signal was transferred to a data acquisition connected to a PC for storing and manipulating the data. The mixture of combustion air and recycled

flue gases was measured by standard orifice plate manufactured according to British standard (BS1042). The differential pressure was measured by a variable angle inclined manometer of range 250 mm H₂O. A mixture of flue gases and pure oxygen was used instead of atmospheric air to reduce nitrogen introduced to combustion process so that the total mass of oxidizer and mass of oxygen were held constant. To do so, a pure oxygen is injected in oxidizer line. The pressure of oxygen was regulated and its flow was controlled and measured using calibrated rotameter.

For flame temperature and appearance purposes, another specially designed combustor of a square cross section was used. One longitudinal side of the combustor was made of a heat-resistant clear glass (glass window) and five ports on the opposite side was used to introduce R-Type thermocouples for flame temperature measuring. The flame photos were taken by the use of a professional digital camera (CANON). The shutter speed, the ISO and the aperture were adjusted according to the flame luminosity in each FGR%. The camera position with respect to the furnace glass window was kept constant to keep the flame scale constant for comparability. The FGR, exhaust gas and cooling water temperatures were measured by K-Type thermocouples as indicated in Fig. 1.

The flue gas analysis was carried out for the measurement of the species O₂, CO, CO₂, UHC and NO concentrations. The sampling was achieved using multi-hole stainless steel probe, which can withdraw an average gas sample all over the whole chimney diameter. The sampling pump was 100% oil-free diaphragm pump and gas sample was filtered with a sequence of filters of different sizes (coarse, fine and very fine). To get a dry gas analysis, a sample chiller was used to separate water vapor from the sample before entering instruments. The analytical instrumentation included a paramagnetic analyzer for O₂ measurements, non-dispersive infrared gas analyzers for CO, CO₂ and NO measurements and a flame ionization detector for UHC measurements. Zero and span standard gases were used periodically for the calibration of the instruments.

The accuracy of each measuring was as follows: fuel flow $\pm 2.5\%$, air flow $\pm 1\%$, oxygen flow $\pm 2\%$, oxidizer flow $\pm (0.6 - 1) \%$, temperature 1.5°C , CO₂, CO, UHC $\pm 1\%$, NO, O₂ $\pm 0.1\%$.

The combustion tests with the flue gas recirculation were carried out based on the lean equivalence ratios from 0.95 down to 0.7 as a typical value considering industrial applications.

The recycling ratio R is defined as the ratio between the mass of flue gas being recycled and the total flue gas mass flow

$$R = \text{recycled flue gas} / \text{total flue gas} \quad (\text{Equation 1})$$

For the purpose of a comparison, the emission concentrations measured in the flue gas were corrected to 3% oxygen by applying the sequence specified in [19] and the next equation [20]:

$$X_{i,3\%} = [(21 - 3) / (21 - X_{O_2})] X_i \quad (\text{Equation 2})$$

where; X_i is the pollutant concentration expressed on a dry volume basis, $X_{i,3\%}$ is the concentration corrected to 3% O₂ and X_{O_2} is the percent dry oxygen concentration in the actual conditions. The results are compared and the difference was negligible.

The experimental work was started by heating up the facility with the full power of 20 kW and taking flue gas recirculation gradually into use. When the chamber had reached a sufficient temperature level, the atmospheric air was reduced and equivalent mass of FGR and pure oxygen was adjusted to the recirculation rate R to keep constant equivalence ratio.

Table 1 summarizes oxidizer conditions for experimental tests at different equivalence ratios.

Table 1. Summary of combustion conditions

Φ	*O ₂ %	FGR % (mass)	Air \dot{m} kg/h	Φ	*O ₂ %	FGR % (mass)	Air \dot{m} kg/h
0.7	0	0	35.9	0.8	0	0	31.4
	13.6	10	30.9		13.7	10	27.1
	27.3	20	26.1		27.4	20	22.8
	40.9	30	21.2		41.2	30	18.5
	54.5	40	16.3		54.9	40	14.2
	68.2	50	11.4		68.6	50	9.9
	81.8	60	6.5		82.3	60	5.6
	100	73.4	0		100	72.9	0
0.9	0	0	27.9	0.95	0	0	26.4
	13.8	10	24.1		13.8	10	22.8
	27.6	20	20.2		27.7	20	19.1
	41.4	30	16.3		41.5	30	15.4
	55.2	40	12.5		55.4	40	11.8
	69.0	50	8.6		69.2	50	8.1
	82.8	60	4.8		83.1	60	4.5
	100	72.4	0		100	72.2	0

*O₂% is defined as the ratio of pure oxygen to the total combustion oxygen

RESULTS AND DISCUSSION

In this study a series of experiments have been made for the investigation of the effect of oxygenated FGR on the flame appearance and emissions.

As shown in table 1, four different cases were considered; the first condition in each case was considered as the reference one with typical air as oxidizer. For each case the mixture of FGR and pure oxygen was increased gradually to the condition of atmospheric air was not used.

Since every case uses the same amount of O₂ flow rate in order to satisfy constant equivalence ratio, therefore, the flame temperature of individual case was expected to show the same temperature behavior as. In Fig. 2 temperature distributions for case 0.8 equivalence ratio is presented. The temperatures were measured at the center plane of the flame. A slightly change in temperature distribution from each condition to other can be observed. The returned flue gas contains unburned vaporized fuel and hydrocarbons, if any, from the previous combustion process even if some traces. The combustion of radicals and hydrocarbon vapors enhances combustion process and increases the reaction rate which, in turn, increase the flame temperature. With increasing recirculation rate the amount of CO₂ in the combustion zone consequently increase. Over 40 % FGR the reduction of temperature caused by radiation heat transfer was occurred where the increase of CO₂ mass decrease flame temperature by the increase of radiation heat loss.

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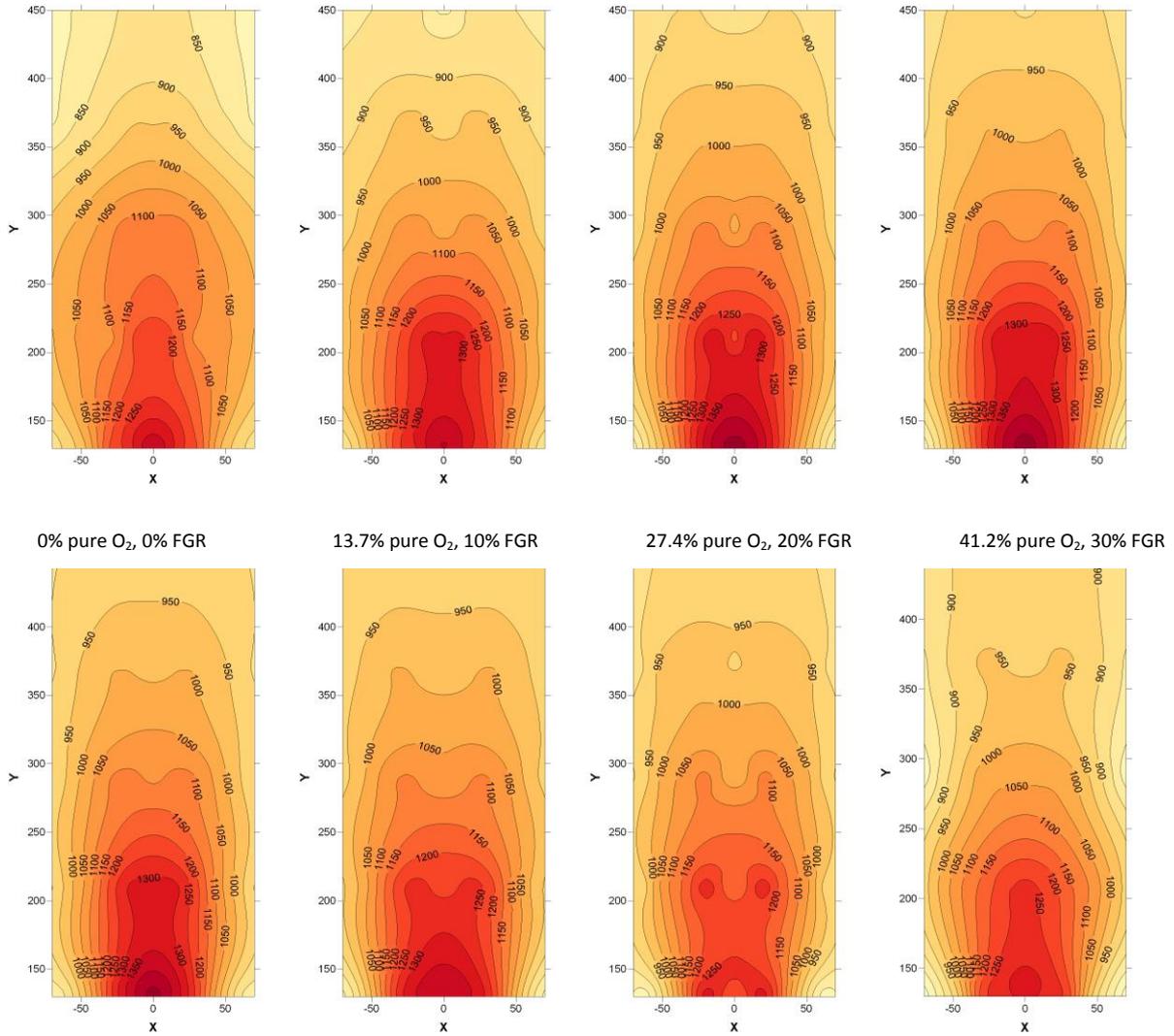


Fig. 2. Temperature distributions at flame center plane for equivalence ratio of 0.8 and different FGR.

The C_p of CO_2 and N_2 are dependent on flame temperature since the specific heat of N_2 is higher than CO_2 below about 600 K and vice versa above 600 K as shown in Fig. 3.

It was observed that the same mass flow rate of CO_2 instead of N_2 have the same heat capacity since the lower C_p value for the CO_2 relative to N_2 at lower temperatures cancels the effect of the higher C_p value at higher temperatures over the examine range of flame temperatures.

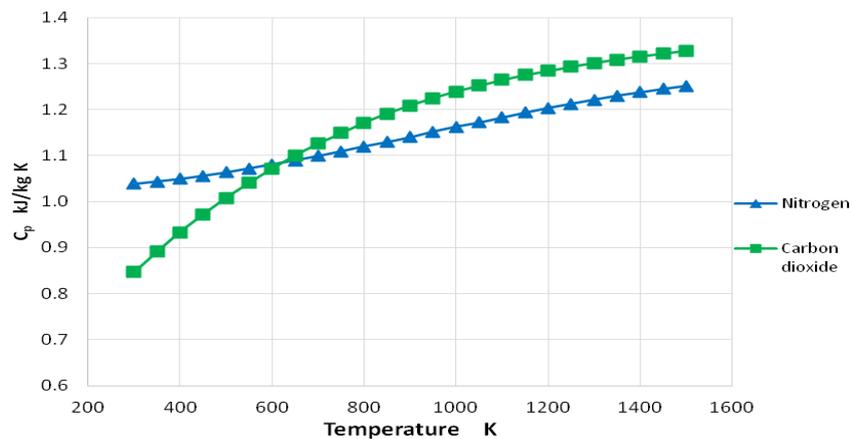


Fig. 3. Specific heat of CO_2 and N_2 as a function of temperature.

Figure 4 shows the flame images of flame at equivalence ratio 0.8 for various recycling ratios. The recycling ratios are indicated in table 1 with relevant values of pure oxygen percentages. It can be observed that for different FGR ratios, the flame length and gases volume have the same general appearance and seem to have the main characteristics of reference condition i.e. atmospheric air as oxidizer. Thus, when the application of FGR with pure oxygen is required, no special design for combustion chamber is needed where the total input mass for all conditions was kept constant. Also, no special materials are required to withstand elevated temperature where hot spots were avoided in this method of application.

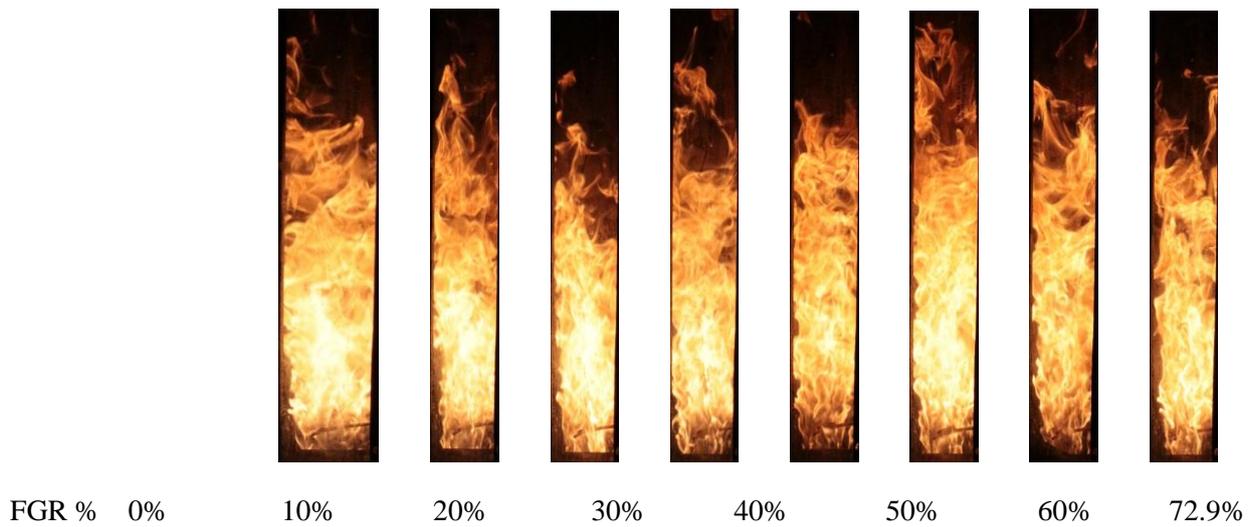


Fig. 4. Flame appearance at different FGR% and Φ 0.8.

The effect of increased temperature and radiation heat loss on system performance can be observed in Fig. 5 which represents the cooling load as a percentage of the fuel heating value which was transferred to the cooling water for different recycling and equivalence ratios. The overall system performance was increased with FGR%. At lower FGR% the convective heat transfer was enhanced by the elevated temperatures with the same mass flow rate. Nevertheless at higher FGR% the temperatures were reduced, the radiative heat transfer was enhanced due to the increased presence of CO_2 particles.

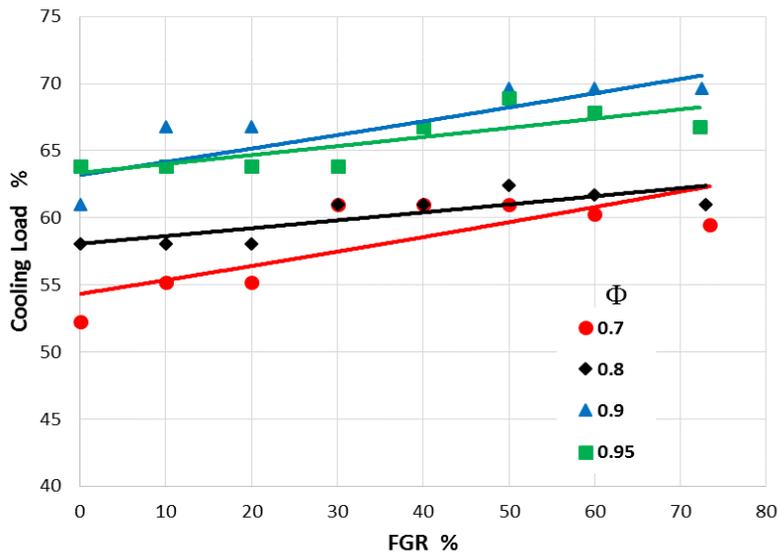


Fig. 5. Cooling load at different recycling and equivalence ratios.

The effect of FGR on NO emission (reduced to 3% O₂ in the combustion products) can be seen in Fig. 6 for different equivalent ratios. The tests were carried out from 0% FGR to the percentage of FGR at which no atmospheric air was introduced to combustion process. For each case the equivalence ratio was unchanged by keeping constant mass of oxidizer with 21% (Vol.) oxygen content.

It can be observed that NO concentration increases with FGR until certain value after which goes into decrease. The increase of NO was due to higher temperature caused by recirculated radicals and unburned vapors as previously discussed. Then the effect of CO₂ radiation heat loss became dominant and NO decreased. In order to achieve a low NO emission by oxygenated flue gas recirculation, more percentages of flue gas should be recycled, and thus more driving power is necessarily consumed. For instant, at equivalence ratio 0.8 the NO has the same value of reference condition at about 40% FGR which is compatible with temperature distributions observed before in Fig. 2. The value of FGR% at which peak NO took-place increases with leaner conditions. The main factor of NO reduction is the lower N₂ partial pressure in oxidizer which inhibits the formation of thermal NO. At the maximum value of FGR% the NO approaches to zero and the recorded values were due to leakage effect.

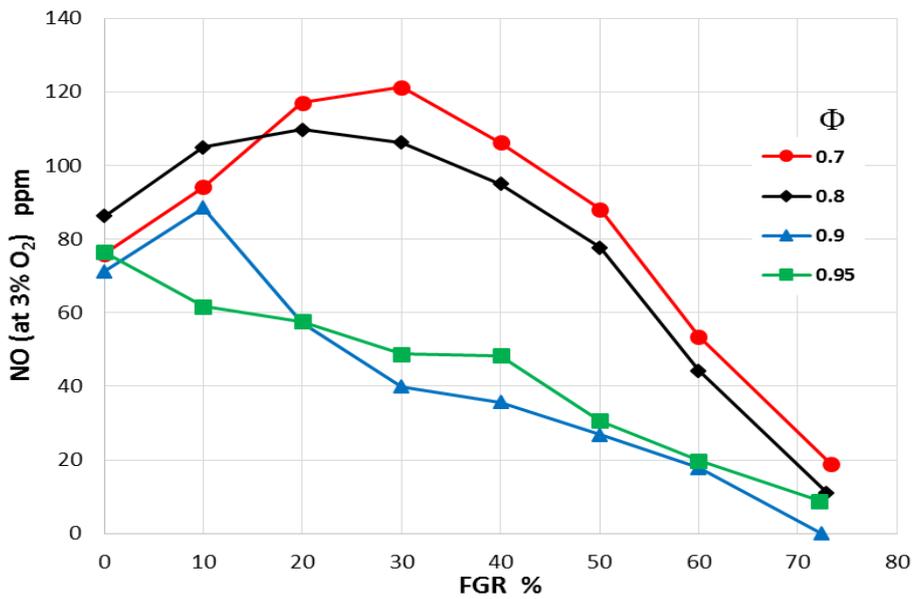


Fig. 6. Effect of FGR on NO emission at different equivalence ratios.

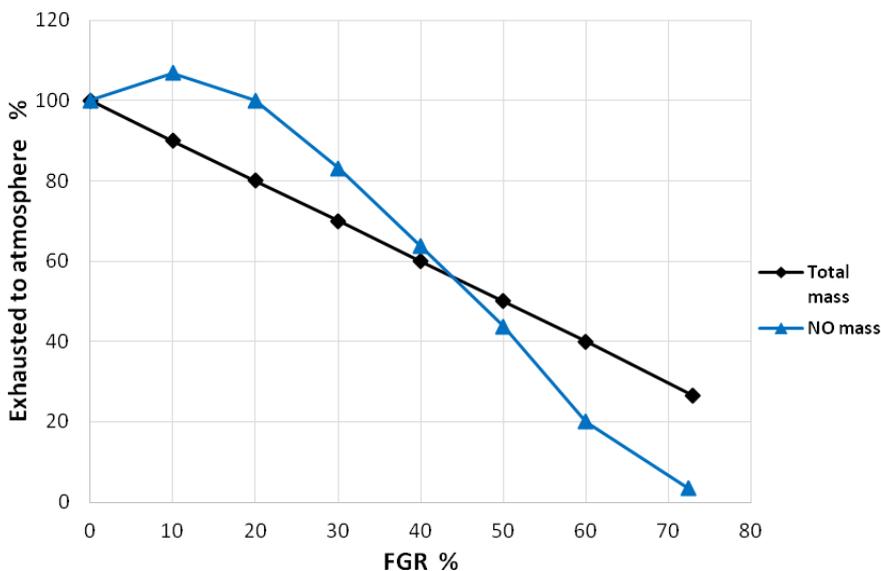


Fig. 7. The relative values of mass and NO exhausted to atmosphere for equivalence ratio 0.8.

Despite increase of NO concentration at the low range of FGR, the actual value emitted to atmosphere was less than that indicated in Fig. 5 and the peak took-place at lower FGR%. This fallback was due to the mass reduction of flue gases exhausted to atmosphere. In Fig. 7 the exhausted mass to atmosphere and corresponding value of NO are represented as a function of FGR% and relative to reference condition value for equivalence ratio 0.8. In this figure, due to the constant mass of oxidizer, when increasing the recirculated mass the exhausted flue gases (which represent the total mass of combustion product minus mass of FGR) was decreased by the same value. Hence the actual mass of exhausted NO to atmosphere were reduced by the same percentage of mass. For instance, at equivalence ratio 0.8 the peak value of NO was shifted to be at 10% FGR instead of 20 % FGR.

CONCLUSIONS

The effect of FGR on oxy-fuel flame appearance and emissions was experimentally investigated using liquid fuel (kerosene) for different equivalence ratios. The main conclusions of this work are summarized as follows:

1. Recirculation of flue gas in oxy-fuel combustion is a suitable method to reduce NO emissions and increase CO₂ concentration for CCS without special design for combustion chamber or using special materials to withstand elevated temperature where the total input mass is kept constant and hot spots are avoided.
2. The flame temperature shows the same behavior as air combustion condition and lower C_p value at lower temperatures cancels the effect of the higher C_p value at higher temperatures over the examine range of flame temperatures.
3. The heat transfer increases by 5 – 10% with increasing FGR% and the radiative heat transfer is enhanced due to the increased presence of CO₂ particles.
4. The reduction of NO emissions takes-place after certain value of FGR% which increases for leaner conditions. The reduced mass of exhausted flue gas has further reduction effect on NO emissions.

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