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High-efficiency combustion of natural gas with 21-30% oxygen-enriched air

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ABSTRACT

This investigation was aimed at studying the influence of 21-30% oxygen concentration on the heating rate, emissions, temperature distributions, and fuel (natural gas) consumption in the heating and furnace-temperature fixing tests. Increase in the oxygen concentration led to a more rapid heating rate and lesser fuel consumption due to lower levels of the inert gas (N₂). When the oxygen concentration was increased from 21% to 30%, the heating rate was increased by 53.6% in the heating test and the fuel consumption was reduced by 26.1% in the furnace-temperature fixing test. Higher oxygen concentrations yielded higher flame temperature; hence, the NO_x emission increased with increasing oxygen concentration. However, the increase of NO_x emission in the furnace-temperature fixing test was less than that in the heating test. Moreover, the NO_x emission was more sensitive to the excess oxygen at higher oxygen levels. The CO₂ concentration in the flue gas increased linearly with the oxygen concentration. Additionally, the temperature distributions became progressively nonuniform with increasing oxygen concentration ally, the temperature distributions became progressively nonuniform with increasing oxygen concentration because the convective heat transfer coefficient was altered.

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1. Introduction

Recently, the cost of energy production has shown a sharp increase and hence improving the energy efficiency is a very important concern. Oxygen combustion technology is one of the useful energy-saving technologies for combustion systems. In traditional air/fuel combustion, nitrogen is used as the inert gas, which siphons away a part of the energy to form flue gas, which lowers the fuel-utilization efficiency. In contrast, oxygen combustion can overcome this disadvantage because of the lower nitrogen concentration involved. The benefits of oxygen combustion include higher thermal efficiency, lower loss of heat energy through the flue gas, higher available heat, higher turndown ratio, and so on [1]. Generally, oxygen burners are applied in high-temperature environments, such as aluminum-remelting furnaces, incinerators, glass-melting furnaces, and ladle furnaces. The oxygen used in these applications is often pure. The high cost of oxygen production therefore limits the widespread application of oxygen combustion. Moreover, if the existent combustion system were to apply oxygen combustion, the burner and furnace body have to be retrofitted substantially due to potential problems, such as heat transfer, flame structure, and refractory damage. Moreover, the cost of retrofitting is very high. The possible solution is to reduce the oxygen concentration during the application of oxygen combustion to existent conventional air/fuel

burners. Economically, this method can save retrofitting and oxygen costs. However, the characteristics of low-level oxygenenrichment in an air/fuel combustion system have not been studied in detail thus far.

Dalton and Tyndall [2] and Joshi et al. [3] adopted conventional air/fuel burners to carry out oxygen-enriched combustion. They found that air/fuel burners could be adapted to low-level oxygen-enrichment (generally lower than 28% O₂) with no modifications. Qiu and Hayden [4] investigated oxygen-enriched combustion of natural gas in porous ceramic radiant burners, at oxygen concentrations varying between 21% and 28%. The experimental results indicated that the saving in natural gas was about 22% when oxygen concentration was increased to 28%. Czakiert et al. [5] carried out combustion in an oxygen-enriched atmosphere using a circulating fluidized bed combustor. They used brown coal as the fuel, and the oxygen concentration was set as 21%, 40%, and 60%. The experimental results showed that the ratio of carbon conversion increased with oxygen concentration. Moreover, oxygen-enriched conditions promoted an increase in the conversion of sulfur present in the fuel. Tan et al. [6] used downfired vertical combustors to study the oxygen-enriched air and O_2/CO_2 combustion. They concluded that very high levels of NO_x formation are achieved owing to higher furnace-temperature and higher oxygen concentrations in the feed air used in oxygen-enriched air combustion. In O_2/CO_2 combustion, the NO_x formation is suppressed because N₂ is absent in the feed air. Poirier et al. [7] adopted low-NO_x burner to explore the influence of oxygen concentration on fuel consumption and emission. The



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experimental results indicated a linear decrease in fuel consumption with increase in oxygen-enrichment when furnace-temperature was maintained at 1100 °C. NO_x emission was relatively constant when oxygen-enrichment levels were below 60% and the stack oxygen concentrations were maintained at the same level. However, NO_x emission increased with increasing stack oxygen concentration at all oxygen-enrichment levels. Chen and Axelbaum [8] studied the variations of the extinction strain rate under oxygen-enriched conditions both numerically and experimentally. Oxygen-enriched combustion could yield extremely strong flames. They found that the extinction strain rate for a methane flame increased by more than two times as the oxygen concentration was increased to 100%. Bejarano and Levendis [9] observed the behavior of coal char in a drop-tube furnace in oxygen-enriched atmospheres. When the gas temperature was maintained within the range of 1300–1500 K, the average temperatures of the char surface increased from 1600-1800 K in air to ranges of 2100-2300 K and 2300-2400 K in 50% and 100% O₂, respectively. In addition, the combustion durations of chars were reduced from 25-45 ms in air to 8-17 ms and 6-13 ms in 50% and 100% O₂, respectively. Murphy and Shaddix [10] conducted a similar study showing that the ranges of oxygen concentration and gas temperature for the combustion of coal chars were 6-36% and 1320-1800 K, respectively. The experimental and numerical results were similar to those reported by Bejarano and Levendis [9] and showed that as the oxygen concentration increased, the char-combustion temperature was increased whereas the char burnout time was reduced. Kennedy et al. [11] investigated the effects of oxygen concentration on filtration-combustion waves. The equivalence ratios and oxygen concentrations studied were in the ranges 0.1-2% and 10-30%, respectively. They found that the peak combustion temperature decreased with increasing oxygen concentration, whereas the absolute flame-propagation rate increased. Furthermore, the CO emission index decreased with increasing oxygen concentration, except for oxygen concentrations higher than 25% and an equivalence ratio near 1.0, when the reaction zone contracted to an area lower than the characteristic pore diameter. Molina and Shaddix [12] proved that ignition occurred faster at higher oxygen concentrations during both O_2/CO_2 and O_2/N_2 combustions applied for combustion of coal. For the same oxygen concentration, the ignition time for O_2/CO_2 combustion was longer than that for O₂/N₂ combustion. The experiment of Liu and Okazaki [13] yielded the same results.

An important barrier to the use of oxygen-enhanced combustion in combustion systems, such as boilers, especially utility-combustion systems, is the current cost of oxygen production and retrofitting. The possible solution is using low-level oxygenenrichment combustion. Huang et al. [14] had analyzed economical feasibility of replacing traditional combustion into oxygen-enriched one for heating furnace, whose flue gas temperature is 920 °C. The ROI (Return on Investment) is about 5.75 years when the oxygen source is generated by membrane method for 30% of O2. In comparison to the above-mentioned studies, fewer studies have explored the characteristics of low-level oxygen-enrichment combustion. This study has used a gas-fired burner to investigate the effects of oxygen concentrations lower than 21-30%. The experiment is divided into two parts - the heating test and the furnace-temperature fixing test. The measurements included heating rate, emissions, temperature distributions, and fuel consumptions.

2. Experimental setup

Fig. 1 shows a schematic representation of the experimental apparatus and the burner configuration. The dimensions of the

combustion chamber were $30 \times 30 \times 100$ cm. The combustor walls were covered with 25 cm thickness of ceramic fiber, which can resist the high-temperature up to 1600 °C. The burner was the North American 4423-0 gas-fired burner. The flame was ignited by a spark pilot, and the combustion occurred in a nonpremixed mode. Both air and oxygen were let into the gas mixer that had a flowmeter to adjust the oxygen concentration in the combustion chamber. The flue-gas compositions (O2, CO, CO2, and NO) were measured using a Testo 350-XL gas analyzer. The measurement ranges of the gas analyzer were 0-25% for O_2 (with an accuracy of about ±0.2% and a resolution of 0.1%), 0-4800 ppm for CO (with an accuracy of about ±5 ppm within 0–99 ppm and ±5% within 100–4800 ppm and a resolution of 1 ppm), 0–25% for CO₂ (with an accuracy of about ±1% and a resolution of 0.01%), and 0-2000 ppm for NO_x (with an accuracy of about ±5 ppm within 0– 99 ppm and ±5% within 100–2000 ppm and a resolution of 1 ppm). The concentrations of species were expressed on a dryvolume basis. For an appropriate comparison, the concentrations of CO, CO₂, and NO_x were corrected to 6% O₂ levels, based on the following equation:

$$[S]_{c} = [S]_{m} \times \frac{[O_{2}]_{a} - 6}{[O_{2}]_{a} - [O_{2}]_{m}},$$
(1)

where $[S]_c$ is the CO, CO₂, or NO concentration corrected to 6% O₂; $[S]_m$ is the measured CO, CO₂, or NO concentration; $[O_2]_a$ is the O₂ concentration of the combustion air supplied; and $[O_2]_m$ is the measured O₂ concentration in the flue gas. Temperature distributions in the combustion chamber at various O₂ concentrations were measured by thermocouples. The measured points totaled thirty, and the location of each point is shown in Fig. 2. The temperature distributions within the combustion chamber were measured by inconelcoated K-type thermocouples. All thermocouples are 0.5 mm diameter of wires and shield by 1.6 mm diameter of inconel tube. The accuracy of the K-type thermocouple provided by the Omega Engineering Inc., was 0.75% of the full scale. The radiative heat loss from the thermocouples was not corrected in the present study. A Fluke 2680A data-acquisition system recorded the temperature values at a recording frequency of 1 Hz. Natural gas, with a heating value of 42 MJ/m³, supplied by the Chinese Petroleum Corporation, was used as the fuel.

In the industry application, the usages of combustion are generally on the heating and/or heat treatment. In heating process, the requirement may be fast heating, such as ladle furnace, or high-temperature, such as glass-melting furnace. In many heat treatment procedures, they are usually limited at specific temperature to perform. Due to these, we select these two cases in this experimental study. For the heating test, the target heating temperature was 1200 °C, and the flow rate of natural gas was maintained at 36 L/min. When the furnace-temperature reached 1200 °C, the experiment was stopped, and the elapsed time was recorded. For the furnace-temperature fixing test, the flow rate of natural gas was changed and the furnace-temperature was maintained at 1220 ± 10 °C under different oxygen concentrations. The experiment was started when the furnace-temperature was maintained at approximately 1220 ± 10 °C for a period of at least 90 min, and the duration of the experiment was 90 min. After the heating and furnace-temperature fixing tests, the oxygen and airflow rates were first adjusted to obtain the desired oxygen concentration and then the experiments were started. In this study, five oxygen concentrations were selected for the experiments - 21%, 24%, 26%, 28%, and 30%. Moreover, the equivalence ratios corresponding to the oxygen concentrations from 21% to 30% are maintained in the neighborhood of 0.952.



Fig. 1. Schematic diagram of the experimental apparatus and burner configuration.



Fig. 2. The measuring positions of thermocouples.

3. Results and discussion

In this section, the effect of oxygen concentration on the combustion characteristics, including heating rate, temperature distributions, emissions, and fuel consumption, are described. In the experiments, the furnace-temperature indicates the value measured by the furthest thermocouple.

3.1. The effects of oxygen concentration as determined by the heating test

Fig. 3 displays the time elapsed for heating the natural gas fuel to 1200 °C at different oxygen concentrations. As shown in Fig. 2, the time elapsed was decreased as the oxygen concentration increased. This is due to the reduction in the inert gas, nitrogen, as the oxygen concentration increased. Hence, the energy availability is raised. The definition of energy availability (η) is as follows:

$$\eta = \frac{E_{\rm in} - E_{\rm loss}}{E_{\rm in}} \tag{2}$$



Fig. 3. Elapsed time of heating to 1200 °C at different oxygen concentration.

where E_{in} is the energy input and E_{loss} the energy loss. The major energy loss is from flue gas. Diluting N₂ concentration can reduce energy loss by flue gas apparently so that the energy availability can be raised, i.e. saving energy. Because of this, the combustion heat was more efficient in increasing the furnace-temperature. Moreover, the magnitude of decrease in the elapsed time was more obvious at higher oxygen concentrations. For example, the elapsed time for 24% O_2 was 83.3% of the time recorded at 21% O_2 (4212 s for 21% O₂ and 3507 s for 24% O₂); however, for 30% O₂, the elapsed time was only 46.4% of that at 21% O₂ (1954 s for 30% O₂). In other words, using combustion air containing 30% O₂ in the heating test could save 53.6% of the fuel compared to using combustion air with 21% O₂. This shortening of the heating time implied savings in terms of fuel. Furthermore, the heating rate was comparable for all O₂ concentrations when the furnace-temperature was lower than 700 °C. This phenomenon indicated that the advantage of oxygenenriched combustion was apparent only in the high-temperature environment during the heating process.

Regarding emissions, the CO concentration and NO_x emission of the flue gas, corrected to 6% O₂, are shown in Fig. 4. The NO_x emission was defined in this study as the sum of NO and NO₂ emissions. According to the experimental results, the CO concentration was maintained at a low-level (<30 ppm) for all O₂ concentrations. The combustion efficiency (*C.E.*) is defined as follows:

$$C.E. = \frac{[CO_2]}{[CO_2] + [CO]} \times 100\%$$
(3)

where [CO₂] is volume concentration (dry) of CO₂ [CO] is volume concentration (dry) of CO. Based on Eq. (3), the combustion efficiencies could achieve above 99.9% in this study. This indicated that excellent combustion efficiency was achieved. However, the NO_x emission increased sharply from 30.95 to 137.29 mg/MJ as the O₂ concentration increased from 21% to 30% O₂, as shown in Fig. 4. However, the maximum furnace-temperature increased to about 50 °C with an increase in O₂ concentration from 21% to 30% (Fig. 5). The variation in temperature was relatively minor, but NO_x variations reached levels greater by more than 4.4 times. Generally, the maximum proportion of NO_x produced during combustion was thermal NO_x . The production of thermal NO_x was directly associated with the temperature and the oxygen concentration. Moreover, the thermal NO_x showed exponential growth with temperature. Due to this, even a minor variation in temperature caused a huge increase in NO_x emission. In addition, it was found that the NO_x emission was more sensitive to the excess oxygen prevalent at higher oxygen concentrations during the experimental

Fig. 4. Variation of CO and NO_x emission at different oxygen concentration in the heating test.



process. Increase in excess oxygen by even a small amount caused a sharp increase in NO_x emission. Because of the effects of temperature and excess oxygen, NO_x emission increased obviously with the oxygen concentration.

Fig. 5 compares the trends of CO₂ concentration (corrected to 6% O_2) and the maximum furnace-temperature (T_{max}) at different O_2 concentrations. As shown in the figure, the CO₂ concentration increased almost linearly with the oxygen concentration. However, the measured values of CO concentration are very low in all cases, indicating that the complete combustion is reached for all cases. So the CO₂ formations are nearly the same for all cases. Because the N_2 concentration in the combustion air was decreased as the O_2 concentration was increased, the N2 concentration was lowered in the flue gas also, causing the CO₂ concentration in the flue gas to increase although the magnitude of CO2 emission remained the same. This phenomenon can reduce the post-processing cost and improve CO₂ capture efficiency. In terms of the maximum furnace-temperature, the higher the oxygen concentration, the higher was the maximum furnace-temperature. For example, the maximum furnace-temperature was 1285 °C at 21% O₂, and it increased to 1337 °C at 30% O₂.

Temperature distributions for different O_2 concentrations during the process of heating the sample to 1200 °C are displayed in Fig. 6. It is evident from the figure that temperature distributions were uniform at points away from the burner. However, the temperature rose gradually near the burner region as the oxygen concentration increased. For example, the temperature was about 1260 °C for 21% of O_2 (Fig. 6a), whereas for 30% of O_2 , it rose to about 1300 °C (Fig. 6e). Moreover, the temperature variations became greater near the region of the burner, with the temperature gradient becoming progressively greater (higher contour density) as the oxygen concentration increased.

On the basis of the results of the heating test, it can be summarized that the heating rate, the maximum furnace-temperature, and the NO_x emission and CO_2 concentration were increased with the oxygen concentration.

3.2. The effects of oxygen concentration in the furnace-temperature fixing test

Fig. 7 illustrates the variations of CO_2 concentration and rates of fuel flow (i.e. fuel consumption) at different oxygen concentrations in the furnace-temperature fixing test. The trend of CO_2 concentration was the same as that in the heating test – that is CO_2 concentration increased linearly with oxygen concentration. In addition,







Fig. 6. Temperature distributions of heating to 1200 °C for (a) 21% O₂, (b) 24% O₂, (c) 26% O₂, (d) 28% O₂ and (e) 30% O₂ in the heating test.

increasing the oxygen concentration could in fact save energy, as shown in Fig. 7. The fuel consumption decreased from 23 L/min at 21% O₂ to 17 L/min at 30% O₂ at a constant furnace-temperature of 1220 ± 10 °C. This implied that 26.1% of fuel could be saved when the oxygen concentration was increased to 30%. Excluding the lower levels of N₂ contained within, the radiative heat transfer was enhanced due to higher concentrations of CO₂ and H₂O when the O₂ concentration was increased. Because CO₂ and H₂O are good radiation emitters, the total gas emissivity was increased when the oxygen-enrichment level was upgraded; in turn, caused the radiation intensity to become stronger. In summary, increasing the oxygen concentration could upgrade the availability of energy. In addition, the fuel consumption decreased almost linearly with increasing O₂-enrichment level at constant furnace-temperature. This trend was the same as the experimental results of Poirier et al. [7].

Fig. 8 shows the variations of CO concentration and NO_x emission, corrected to 6% O₂, at different oxygen concentrations in the furnace-temperature fixing test. From this figure, the CO concentration can be observed to be lower than 6 ppm for all O₂ concentrations. This indicated that incomplete combustion did not occur. The trend of NO_x emission was also assessed. In the heating test, the NO_x emission increased dramatically as the oxygen concentration was enhanced. Similarly, NO_x emission in the furnace-temperature fixing test showed an increasing trend; i.e., NO_x concentration increased with oxygen concentration, but the NO_x concentration was much less than that in the heating test at the same oxygen concentration. For example, at 30% O₂, the



Fig. 7. Variations of CO₂ concentration and fuel flow rate at different oxygen concentration in the furnace-temperature fixing test.



Fig. 8. Variations of CO concentration and NO_x emission at different oxygen concentration in the furnace-temperature fixing test.

 NO_x concentration was 545 and 95 ppm in the heating test and the furnace-temperature fixing test, respectively. Moreover, the NO_x concentration increased 6.33 times (from 86 to 545 ppm) as the oxygen concentration was increased from 21% to 30% in the heating test, but it only increased 2.11 times (from 45 to 95 ppm) in the furnace-temperature fixing test. As mentioned in Section 3.1, the production of thermal NO_x was affected by temperature severely. The maximum furnace-temperatures in the furnace-temperature fixing test, as shown in Fig. 9, were lower than those in the heating test (Fig. 5) for all oxygen concentrations. At 30% O₂, the maximum furnace-temperature in the furnace-temperature fixing test was 1291 °C, whereas it was 1337 °C in the heating test. Furthermore, the increases of maximum furnace-temperatures in the heating and the furnace-temperature fixing tests with an increase from 21% to 30% O₂ were 52 and 26 °C, respectively. The reason is that the energy input in the heating test is greater than that in the furnace-temperature fixing test. Consequently, the increases in the concentration of NO_x were relatively higher for the heating test. Although the NO_x concentration increased with oxygen concentration, the total NO_x emission did not show an increase similar to that of the concentration because the magnitude of flue gas also decreased when the oxygen concentration increased. Because the total NO_x emission is a product of the volume concentration of NO_x and the flue



Fig. 9. Variation of maximum furnace-temperature at different oxygen concentration in the furnace-temperature fixing test.

gas volume flow rate, these two effects - increase in NO_x concentration and decrease in amounts of flue gas - competed with each other. In addition, there was a peculiar value of NO_x emission for 24% O_2 ; i.e., the NO_x emission was lower than 21% O_2 . The possible reason was that the emission of thermal NO_x was affected not only by temperature, but also by excess oxygen. The maximum furnace-temperature at 24% O_2 was higher than that at 21% O_2 (1265 °C for 21% O_2 and 1268 °C for 24% O_2); however, the excess oxygen was lower at 24% O₂ (1.85% for 21% O₂ and 1.30% for 24% O₂). Increasing the furnace-temperature caused an increase in the NO_x emission, but reducing the excess oxygen caused NO_x emission to decrease. Although the effect of the furnace-temperature was greater than that of excess oxygen (the effect of temperature was exponential), as long as the hot spots (the zones with a concurrent occurrence of relatively high-temperature and oxygen concentration) were less in number, the quantity of NO_v emission was reduced. Thus, the NO_x emission under 24% O₂ was less than that at 21% O₂.

The temperature distributions at different oxygen concentrations in the furnace-temperature fixing test are shown in Fig. 10. From the figure, the higher temperature zones were observed to be concentrated near the burner region, and their area was increased with the oxygen concentration. Moreover, when the oxygen concentration was high, the uniformity of the temperature field was disturbed. The reason was that the convective heat transfer was altered, being directly associated with the velocity and flow pattern of the fluid. The combustion needed less air when the oxygen concentration was increased. Moreover, the fuel consumption decreased with increasing oxygen concentration that caused the required amount of combustion air to be less. Thus, the velocity and flow pattern of the fluid could be inferred to change obviously. These two effects reinforced each other. Therefore, the temperature distribution became nonuniform as the oxygen concentration increased. Comparing with Fig. 6, the uniformity of temperature was worse for furnace-temperature fixing test. The fuel flow rate was maintained constant for the heating test, so its required combustion air is less than that of furnace-temperature fixing test.

Summing up, based on the experimental results of the furnacetemperature fixing test, the fuel consumption decreased and the NO_x emission in the flue gas increased with increasing oxygen concentration. Moreover, the temperature distribution was nonuniform with increasing oxygen concentrations. Finally, the NO_x emission can be controlled at low levels by reducing both the excess oxygen and the furnace-temperature.



Fig. 10. Temperature distributions for (a) 21% O₂, (b) 24% O₂, (c) 26% O₂, (d) 28% O₂ and (e) 30% O₂ in the furnace-temperature fixing test.

4. Conclusions

In the present study, the heating and furnace-temperature fixing tests were adopted to investigate the effects of oxygen concentrations in the range of 21–30%. The influences of oxygen concentration on the heating rate, emissions, temperature distributions, and fuel consumption were examined. The general conclusions drawn from the results of this work were as follows:

- 1. The heating rate was rapid owing to the reduced levels of combustion air as the oxygen concentration was increased. Compared to that at 21% O₂, the time elapsed for heating to 1200 °C was only 46.4% for air with 30% O₂. However, the difference between the heating rates was small when the target heating temperature was lower than 700 °C.
- 2. The NO_x emission increased sharply due to the higher furnace-temperature as oxygen concentration increased in the heating test. The NO_x emission was increased by 4.4 times when the oxygen concentration was increased from 21% to 30%.
- 3. The CO₂ concentration increased almost linearly with the oxygen concentration during both the heating and the furnacetemperature fixing tests.
- 4. For maintaining the furnace-temperature, the fuel efficiency and radiative heat transfer showed increases with increasing oxygen concentration, so that the fuel consumption decreased as oxygen concentration increased. The fuel consumption at $30\% O_2$ was reduced by 26.1%, compared with that at 21% O_2 when the furnace-temperature was maintained at 1220 ± 10 °C.

- 5. The trend of NO_x emission in the furnace-temperature fixing test was the same as that in the heating test; however, the NO_x emission was much less than that in the heating test at the same oxygen concentration owing to the high furnace-temperature, fuel consumption and flue gas flow rate.
- Increasing the oxygen concentration led to nonuniform temperature distributions because the convective heat transfer coefficient was changed.

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