High Efficiency Furnace with Oxy-Fuel Combustion and Zero-Emission by CO2 Recovery

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Keywords: 1. Oxy-fuel combustion; 2. Energy Conservation; 3. Conventional Equipments; 4. NOx; 5. CO2 Recovery.

1 Background

It is necessary to reduce CO2 emissions against global warming, and the activities are expanding all over the world.

In Japan, approximately 85% of primary energy consumption is reliant on non-renewable fossil fuels. And more than 30% of fossil fuels are used in industrial boilers, industrial furnaces and power generation stations.

Natural gas is considered as an environmentally friendly clean fuel, offering important environmental benefits when compared to other fossil fuels. Among fossil fuels, natural gas produces relatively small amount of CO2 per unit calorific value. Furthermore, the superior environmental qualities over coal or oil are that emissions of sulphur dioxide are negligible or that the level of nitrous oxide emissions is lower. So, natural gas will play an important role in meeting our nation's future energy needs.

We, Tokyo Gas Co., Ltd. has contributed to reduce CO2 emissions by exchange fuels that have large CO2 emissions coefficients such as heavy oil and coal to natural gas. And to develop and introduce high efficiency combustion instruments is also our significant environmental policies of CO2 emissions reduction.

A trial calculation made by IPCC (Intergovernmental Panel on Climate Change) requires us to reduce the amount of CO2 down to 50% by the year 2050. Natural gas produces more CO2 than nuclear power or renewable energy such as solar energy. Therefore, it is necessary to take measures to reduce CO2 emissions in order that city gas may become mainstay energy in the future.

We have to think about not only the problem of CO2 emission reduction but tight supply-demand situation in near future and jump in natural gas prices as social responsibility of Tokyo Gas.

Fossil fuel, such as natural gas and oil is well used in the various industrial process and the energy savings on these will result in big effects in reducing of global warming gases. For example, about 10 % primary energy demanded in Japan is used in steel industry. Therefore the energy saving and environment protection technologies should be developed and then they should be applied as soon as possible in the practical fields.
2 Objectives of the paper

In these situations, we have attempted to develop the oxy-fuel combustion system which realize to improve efficiency of furnace and to recover CO2 from its exhaust gas.

Oxy-fuel combustion is the process of burning a fuel using pure oxygen instead of air as the primary oxidant. When the oxygen concentration is raised above the 20.9% present in combustion air, the air is said to be oxygen-enriched. And the chemical reaction between fuels and oxygen-enriched air is called oxygen-enriched combustion.

The features of oxy-fuel combustion are;

1. Oxy-fuel combustion produces approximately 75% less flue gas than air fuel combustion.
2. Because the exhaust gas volume is reduced, less heat is lost in the exhaust gas.
3. Higher flame temperatures are possible. Because nitrogen component of air is not heated.
4. The exhaust gas is consisting primarily of CO2 and H2O, suitable for sequestration.
5. Nitrogen from air is not allowed in, thermal NOx production is greatly reduced.

Thus, oxy-fuel combustion has these advantages, but there were some subjects or problems to be solved due to the following reasons.

1. Firing with pure oxygen would result in too high flame temperature, so inadaptable with burners for air fuel combustion.
2. Reduced flue gas volume will decrease in efficiency of convective heat transfer and make temperature distribution in furnace worse.
3. The invasion of the external air into the furnace and high flame temperature produces large amount of NOx.
4. It costs a lot to produce oxygen.

The oxygen combustion technology is not applied to equipment such as industrial furnaces and boilers though it makes a great profit, because of the above-mentioned problem. Therefore, we developed the application of the oxygen combustion technology to existing equipments. At the same time, we tried to develop and examine to solve a technical problem such as increase of NOx formation or greatly decrease of heat transfer by convection.

In the near future, it is predicted that oxy-fuel combustion system will benefit economically by virtue of a jump in fossil fuels prices or cost reduction of oxygen production.

The justification for using oxy-fuel combustion is to produce a CO2 rich flue gas ready for sequestration. Oxy-fuel combustion has significant advantages over traditional air-fired plants. There is some possibility of applying CCS (CO2 Capture and Storage) because this system can recover CO2 simply.
3 Development

a. About Characteristics of Oxy-Fuel Combustion

Combustion is the chemical reaction between fuel and oxygen. The vast majority of combustion processes use air as an oxidizer in combustion with a fuel such as natural gas, fuel oil, propane, other hydrocarbons and the like. Air is the usual source of oxygen for industrial furnaces or boilers. While readily available for free, air is only about 21% oxygen and 79% nitrogen. Hot exhaust gases represent the largest source for losses in most industrial furnaces. It is recognized that pre-heating the combustion air with heat exchanger provides the potential for energy savings. Since nitrogen does not contribute to the combustion process, it introduces a major source of inefficiency to the combustion process. The nitrogen in the air increases substantially the mass of the flue gas and the heat losses up the industrial furnaces or the boiler stack. Some nitrogen is converted to oxides of nitrogen, which is a major source of air pollution, and contributes to the formation of ozone and photochemical smog.

Oxy-fuel combustion is the process of burning a fuel using pure oxygen instead of air as the primary oxidant. Here are the characteristics of oxy-fuel combustion.

1. Lower volume production of exhaust gas

In case of air fuel combustion, it goes without saying that the large part of exhaust gas consist of nitrogen heated up to high temperature after combustion. It is also known that the performance of many air-fuel combustion processes can be improved by enriching the combustion air with oxygen. Replacing air with oxygen drops volume of exhaust gas and heat loss by 75% in the flue gas from air-fuel combustion. And it is resulting in an overall improvement in efficiency. (Figure.1, Figure.2).

2. Achievement of higher flame temperature

The adiabatic flame temperature of air-fuel combustion is approximately 1950°C. Enrichment of the combustion air increases both the flame temperature and the thermal efficiency abruptly. In oxy-fuel combustion, the adiabatic flame temperature reaches approximately 2800°C. (Figure.3)

3. Improvement of energy efficiency

Enrichment of the combustion air increases both the flame temperature and the thermal efficiency while the flue gas volume decreases as the oxygen concentration in the air or oxidizer increases. Relation between oxygen-enriched rate and fuel saving rate is shown in Figure.1. A higher temperature of flue gas is evaluated the efficiency of energy conservation. Thus, It means that the higher temperature of flue gas, the lower fuel consumption. The concentration increase of CO2 and H2O in the flue gas increases efficiency of heat transfer. The benefits of oxygen-enriched or oxy-fuel combustion in conjunction with high flame radiation substantially increases the thermal efficiency of a furnace.

4. Possibility of capture CO2

Oxy-fuel combustion produces a nitrogen free exhaust gas with water vapour and a high concentration of CO2 as its main components. This makes it easy to further concentrate the exhaust gas to an almost pure stream of CO2. (Figure.4)

5. Achievement of extra low NOx emissions

Relation between oxygen enriched rate and NOx formation is shown Figure.5. Thermal NOx formation on temperature means that the predicted equilibrium NOx from the combustion of a fuel with theoretical flame temperature. It is important to recognize the tendency of NOx formation. Its advantages include a rise in combustion efficiency, due to an increase in partial pressure of oxygen. The reduction of inert, such as nitrogen leads to increase in the adiabatic flame temperature. However, higher flame temperature can also result in higher NOx emissions. When the concentration of oxygen in the combustion air is over 60%, the NOx formation is decreased. Furthermore, oxy-fuel combustion produces no NOx emission theoretically.
Figure 1. Volume of Flue Gas

Figure 2. Variation of Fuel Saving

Figure 3. Adiabatic Flame Temperature
Oxy-fuel combustion is not the latest technology. It is utilized oxy-fuel combustion in particular industrial processes. Here are the methods of oxygen-enriched combustion and oxy-fuel combustion.

1. **Oxygen-Enriched Combustion**

Oxygen-enriched combustion is the combustion art that oxygen enrichment by the addition of high-purity oxygen to the air. Generally, the concentration of oxygen in the oxygen-enriched combustion is about 30%.

It is easy to apply oxy-fuel combustion to conventional burners. And the use of oxygen-enriched combustion in a number of energy-intensive industrial applications has the potential to reduce the amount of heat lost to the atmosphere by three-fourths. Furthermore, the NOx formation of oxygen-enriched combustion is to be kept as same amount as air fuel combustion. But there is a weak point that over 30% of oxygen-enriched rate cause NOx formation. NOx increases greatly because a large amount of nitrogen coexists with oxygen at the same time and the flame temperature rises though the effect of energy conservation is large.
2. Undershot (Under flame) Enrichment Combustion

One of the combustion systems that installs lance that blows pure oxygen under the flame formed with air fuel combustion. While air enrichment increases the flame temperature uniformly, the undershot technique selectively enriches the underside of the conventional flame, thereby concentrating extra heat downward toward the material being heated. However, rise in flame temperature causes a rapid increase in the rate of NOx formation when concentration of oxygen is over 30%. The problem remains as well as the oxygen-enriched combustion.

3. Oxy-Fuel Combustion

Oxy-fuel combustion is the process of burning a fuel using pure oxygen instead of air as the primary oxidant. Since the nitrogen component of air is not heated, fuel consumption is reduced, and higher flame temperatures are possible. Therefore, it is applied to melting furnace such as glass melting tank and cast iron melting furnace. However, it needs exclusive burners to combust fuel with pure oxygen. Moreover, it is important to consider heatproof of equipment so that the adiabatic flame temperature in the oxygen burning may reach near 2800°C.

We tried to develop technologies as fourth oxygen combustion to adapt oxygen-enriched or oxy-fuel combustion to combus natural gas and make efforts to introduce these technologies into the field of industries. Our technical correspondences have effected on melting furnaces that require us high temperature such as glass melting furnaces, ash melting furnaces and cast iron melting furnaces. Oxygen-enriched and oxy-fuel combustion demonstrates the ability to achieve the high temperature which cannot be realized by the air fuel combustion, and it gives great effect of energy conservation.

However, it is not easy to say that the oxygen combustion technology has been received citizenship in industrial processes because of following reasons.

- It costs a lot to produce or buy oxygen.
- There must be a problem in heatproof of equipment with the high temperature flame.
- Oxy-fuel combustion needs exclusive burner.
- Thermal NOx is formed because of high temperature in the oxygen-enriched combustion and the invasion air.
- The exhaust gas volume of oxygen-enriched or oxy-fuel combustion which is smaller than that of air-fuel combustion decreases convective heat transfer and deteriorate the temperature distribution in industrial furnace.

b. The Adaptation of Oxy-Fuel Combustion Technology to Conventional Furnaces

1. Technical problems

To solve technical problems of oxy-fuel combustion, we tried to develop the forced flue gas recirculation system that flue gas is captured from an emissions stack and recirculated to a burner by an induced flue gas recirculation system comprising as fourth oxygen combustion technology. We readjusted matters of concern when we applied oxy-fuel combustion to conventional burners and industrial furnaces.

In many furnaces, the conventional burner combust fuel with air, which of course contains the oxygen needed for the combustion. Such combustion is termed “air-fuel combustion” and burners at which air-fuel combustion occurs are termed “air-fuel burners”. Air-fuel burners cannot use to combust fuel with a gaseous oxidant that contains oxygen in a concentration higher than that of air because of the higher
temperature at which the combustion occurs. Oxy-fuel combustion often requires the use of burners termed “oxy-fuel burners” that are adapted for oxy-fuel combustion, in particular in their ability to withstand the higher combustion temperatures obtained in oxy-fuel combustion. It costs a lot to exchange air-fuel burners to oxy-fuel burners and to take measures to cope with high temperature flame.

Generally, the heat transfer in the industrial furnaces is performed by radiation from flame and convection of flue gas. It is concerned a decline in efficiency of heat transfer and a change for the worse of temperature distribution in the industrial furnaces because of less volume of flue gas caused by oxy-fuel combustion. There is possibility of lowering yield rate and quality of production and we stand to lose everything.

The problem as stated above with utilizing oxygen enrichment in combustion is that there is a dramatic increase in NOx emissions. In most industrial combustion processes, over 90% of the NOx emissions are in the form of nitric oxide or NO. Figure.5 shows that NOx dramatically increases at low levels of oxygen enrichment ranging 30% to 60% which trend has been verified experimentally in numerous tests conducted with enrichment of conventional air-fuel burners.

It is hard to keep furnace pressure positive to avoid the invasion of the external air because of less volume of flue gas caused by oxygen combustion. This problem also might cause the formation of NOx.

There are some other apprehend problems however, the above-mentioned three matters on concern are bottlenecks to adapt technology of oxy-fuel combustion to the industrial furnaces.

2. Proposal system

To solve these problems, we tried to develop the forced flue gas recirculation system which realizes oxy-fuel combustion with conventional air-fuel burners. The point of this system is to supply air with oxygen and recirculated flue gas as combustion air to air-fuel burners. Oxygen and recirculated flue gas are mixed with the air to keep concentration of oxygen as same level as air. It realized to keep the partial pressure of N2 lower than that of air. This combustion air what is called “pseudo-air” consist of CO2, H2O, oxygen, and less or few of N2.

We can determine ourselves the oxygen-enriched rate, air fuel ratio and the volume rate of recirculated flue gas. The conceptual figure of this system is shown in Figure.6.

![Figure.6. Conceptual Figure of the System](image)

The amount of oxygen, air, and the recirculated flue gas are calculated based on the amount of fuel supply and various parameters arbitrarily decided. The volumes of these gases are controlled by control valve adjusting set values. The heat exchanger in Figure.6 can be used as effective use of flue gas such as waste heat boiler.

Oxy-fuel combustion can be realized with the burner for air-fuel combustion by supplying this Pseudo-air to the burner. And, because the amount of flue gas from the burner is as same amount as that of air fuel combustion, the effect of convection heat transfer in the furnace is kept.
An increase in the NOx formation in the oxygen-enriched combustion was feared according to the NOx formation theory shown in Figure 5. It is so hard to keep furnace pressure positive because common furnaces have some openings that it is impossible to prevent the invasion air getting into furnace in fact. Therefore, the counter measure should be taken considering the existence of nitrogen, even though oxy-fuel combustion. The flame temperature which is restrained with the recirculated flue gas decreases of NOx formation by virtue of this system.

The advantages of this system were brought together as follows.

- This system with oxy-fuel combustion will make a significant contribution to the development of technology that can be retrofitted to conventional equipments.
- Because the amount of flue gas from the burner doesn't decrease, convectional heat transfer is kept as same as the air fuel combustion.
- The low-NOx firing combustion is possible according to the control of flame temperature by the recirculated flue gas.
- Air combustion, oxygen-enriched combustion, and oxy-fuel combustion can be switched. And the concentration of oxygen of the entire system is decided up to 21-100% arbitrarily.
- The higher rate of partial pressure of CO2 and H2O, the higher efficiency of radiation heat transfer.
- In the state of oxy-fuel combustion, CO2 can be recovered by 90% or more theoretically.
- Even when the oxygen supply stops, it is easy switching to the air fuel combustion.

c. Experimental Equipments

1. Experimental furnace

Dimension of Furnace : 1500(W)×3150(L)×1200(L) [mm]
Maximum Temperature : 1300 [°C]
To decrease the formation of NOx in high temperature air combustion, we have successfully developed an advanced low NOx combustion technology called FDI (Fuel Direct Injection) in ‘90s. FDI combustion technology reduces thermal NOx substantially for combustion of high preheated air over 1000°C. The principal of its ultra-low NOx combustion is the separate and direct injection at high momentum of combustion air and fuel gas into the furnace. By directly injecting air and fuel, self-induced flue gas recirculation is substantially enhanced, reducing the formation of thermal NOx to a substantially low level.

Applied to a regenerative burner system that utilize high air preheat for fuel saving, the FDI combustion has demonstrated more than 90% NOx reduction. As compared to conventional ones, simple and compact regenerative burners have been developed.

The temperature of recirculated flue gas of our system might reach about 500°C. Combustion of fuels with high temperature pseudo-air could be one of the promising technologies in industrial furnace applications, to reduce CO2 as well as save energy. However, high temperature pseudo-air also might generate great amount of NOx at the same time.

We can use FDI combustion technology when the temperature of pseudo-air is getting high because of recirculated flue gas.

Figure.10 shows the drawing of air fuel burner for FDI combustion. Pseudo-air run through the air fuel burner and fuel gas is supplied to its original nozzle called FDI nozzle.
d. Properties of fuel

We used city gas as fuel of oxygen combustion in whole experiment. Table.1 shows properties of city gas.

Table.1 Properties of City Gas

<table>
<thead>
<tr>
<th>Composition</th>
<th>CH4</th>
<th>Vol%</th>
<th>89.6</th>
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<tr>
<td></td>
<td>C2H6</td>
<td>Vol%</td>
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<td></td>
<td>C3H8</td>
<td>Vol%</td>
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<td></td>
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<td>Vol%</td>
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<tr>
<td></td>
<td>n-C4H10</td>
<td>Vol%</td>
<td>0.7</td>
</tr>
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<td>Total</td>
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<td>100</td>
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<tr>
<td></td>
<td>H</td>
<td>-</td>
<td>4.332</td>
</tr>
<tr>
<td>Carolific Value</td>
<td>HHV</td>
<td>MJ/m3N</td>
<td>45.0</td>
</tr>
<tr>
<td></td>
<td>LHV</td>
<td>MJ/m3N</td>
<td>40.7</td>
</tr>
<tr>
<td>Specific Gravity</td>
<td></td>
<td></td>
<td>1.0</td>
</tr>
<tr>
<td>Theoretical Air</td>
<td></td>
<td></td>
<td>10.7</td>
</tr>
<tr>
<td>Maximum Combustion Speed</td>
<td>cm/s</td>
<td></td>
<td>37.0</td>
</tr>
</tbody>
</table>
4 Results

1. State of combustion at oxygen combustion with FGR (Flue Gas Recirculation)

In general, luminance of the flame increases and temperature of the flame rises when the oxygen-enriched rate is raised. In the state of the pure oxygen combustion, an ashen flame is formed a quite different flame from air combustion is shown. In this experiment, the flame similar to that of air combustion was able to be formed even with the state of oxygen-enriched combustion and the pure oxygen combustion by using pseudo-air. The state of combustion is shown below.

![Figure 11. Flame of Oxygen-Enriched Combustion](image)

![Figure 12. Flame of Oxy-Fuel Combustion](image)

2. CO2 concentration in oxygen combustion

The CO2 concentration in exhaust gas rises as the oxygen enrichment rate goes up. The relation between the oxygen enrichment rate and the CO2 concentration is shown in Figure 13. In this experiment, the excess air ratio was set as 1.05, and the experiment result obtained almost the same result as the theoretical value.

![Figure 13. Concentration of CO2 in Oxy-Fuel Combustion](image)
To accomplish the highest concentration of CO2, we tried to conduct an experiment on oxy-fuel combustion at low air ratio without generating monoxide. The actual value of CO2 concentration was 95.8% (O2: 2.5%, CO: 130ppm) in this experiment.

3. Keeping of convection heat transfer

In oxygen combustion, it is important to keep the amount of exhaust gas from the burner because the convection heat transfer effect is not decreased. By using pseudo-air made by recirculated flue gas, this system almost obtained same amount of flue gas as the air combustion from oxygen-enriched combustion to the oxy-fuel combustion. Figure.14 shows the relation between the oxygen enrichment rate and the amount of flue gas. The amount of flue gas equal with the air combustion can be kept by FGR in oxygen-enriched combustion.

![Figure.14 Volume of Flue Gas](image)

And the actual values are inscribed in Table.2.

<table>
<thead>
<tr>
<th>Oxygen-Enriched Rate %</th>
<th>Gas</th>
<th>Air</th>
<th>O2</th>
<th>FGR *</th>
<th>Flue Gas</th>
<th>FGR less</th>
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<tbody>
<tr>
<td></td>
<td>m3N/h</td>
<td>m3N/h</td>
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<tr>
<td>21</td>
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<td>76.0</td>
<td>314.2</td>
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<td>25.5</td>
<td>122.0</td>
<td>39.0</td>
<td>102.0</td>
<td>301.5</td>
<td>186.5</td>
</tr>
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<td>25.5</td>
<td>79.0</td>
<td>45.6</td>
<td>151.0</td>
<td>301.1</td>
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<td>52.0</td>
<td>48.7</td>
<td>199.0</td>
<td>325.2</td>
<td>126.2</td>
</tr>
<tr>
<td>70</td>
<td>25.5</td>
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<td>199.0</td>
<td>311.5</td>
<td>112.5</td>
</tr>
<tr>
<td>80</td>
<td>25.5</td>
<td>19.0</td>
<td>56.3</td>
<td>201.0</td>
<td>301.8</td>
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</tr>
<tr>
<td>90</td>
<td>25.5</td>
<td>10.0</td>
<td>58.5</td>
<td>198.0</td>
<td>292.0</td>
<td>94.0</td>
</tr>
</tbody>
</table>

* FGR : Flue Gas Recirculation

4. NOx emissions

In the oxy-fuel combustion of the natural gas, the NOx emission is not generated theoretically. We measured concentration of NOx in oxy-fuel combustion and oxygen-enriched combustion. Especially, an increase of the NOx in oxygen-enriched combustion is a matter feared most so far. We tried to prevent an increase of the NOx by controlling the flame temperature by FGR.
4.1. NOx emissions with air combustion

At first, we measured a basic performance on NOx formation of the burner used for the experiment at the air combustion. The experiment burner that is typical nozzle-mix burner burns slowly and NOx emissions are comparatively low. Figure 15 shows the experiment result.

Although NOx concentrations increase when the excess air ratio goes up, it is comparatively low NOx concentration. This experimental data is compared with the data of oxygen-enriched combustion and the pure oxygen combustion as follows.

![Figure 15. Basic Performance of Experimental Burner](image)

4.2. NOx emissions with oxy-fuel combustion

The nitrogen content is not contained in the city gas and oxygen used for experiment from liquid oxygen. In the pure oxygen combustion, the NOx is sure not to be generated theoretically. We measured data with pure oxygen combustion. The measurement result of data concerning the NOx is shown as Table 3.

<table>
<thead>
<tr>
<th>Table 3 Actual Value of NOx Emission with Oxy-Fuel Combustion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Item</td>
</tr>
<tr>
<td>Input : [kW]</td>
</tr>
<tr>
<td>FGR *1 : [m3N/m3N-fuel]</td>
</tr>
<tr>
<td>Furnace Temp. : [°C]</td>
</tr>
<tr>
<td>O2 Concentration *2 : [%]</td>
</tr>
<tr>
<td>CO2 Concentration *2 : [%]</td>
</tr>
<tr>
<td>NOx Concentration *2 : [ppm]</td>
</tr>
<tr>
<td>NOx Concentration *2 : [g/m3N-fuel]</td>
</tr>
<tr>
<td>CO Concentration *2 : [ppm]</td>
</tr>
<tr>
<td>Furnace Press. : [Pa]</td>
</tr>
</tbody>
</table>

*1 : Flue Gas Recirculation  
*2 : dry base
In the pure oxygen combustion, the amount of the flue gas becomes about one fourth. In addition, about two thirds of the exhaust gases are composed by steam. Therefore, the amount of exhaust gas of the pure oxygen combustion with a dry base is about one tenth of the amount of exhaust gas of the air combustion.

The concentration of NOx and the other gas reach a large value compared with the air combustion. 6.7% of the oxygen concentration in the experimental result is about 2.5% for a wet base. It is necessary to describe the weight of the exhaust gas element in parallel in case of oxygen combustion with a lot of vapour in exhaust gas. In the table above, the weight of the NOx per fuel is inscribed. At the furnace temperature 1300°C, the NOx concentration was 21ppm. This value is 7.8ppm in wet base. Moreover, it is about 2ppm when converting it into the same amount of air combustion. It is a very low NOx concentration in high temperature furnace.

In this experiment, we recirculated 8.0m3/Nm3N-fuel of flue gas. However, it does not influence on NOx formation because almost same amount of NOx is measured in low rate of FGR. It is important to shut the invasion air out for control NOx formation in oxy-fuel combustion.

The nitrogen content is sure not to be contained even in the fuel and oxygen, and not to generate the NOx theoretically. The generation of the NOx was observed though it was a minimum amount. This cause is the little air that invaded from piping and the furnace casing, etc. It is thought that air goes into furnace because a part of the recirculation line is a negative pressure though furnace pressure is controlled at +50 Pa.

4.3. NOx emissions with oxygen enrichment combustion

The flame temperature rises greatly because the oxygen enrichment rate goes up, and thermal NOx increases. The amount of NOx formation peaks by the enrichment rate about 60% in the equilibrium calculation. In addition, because the nitrogen content pressure decreases relatively, the NOx concentration starts decreasing oppositely though the flame temperature rises when the oxygen concentration is improved. Figure.3 and Figure.5 show the relation between the oxygen enrichment rate and the adiabatic flame temperature, and show the relation between the oxygen enrichment rate and the amount of the theory equilibrium NOx generation.

We measured the NOx data of the oxygen enrichment in the burner of the experimental equipment. As shown in Figure.3, the adiabatic flame temperature exceeds 2000°C when the oxygen enrichment rate exceeds 30%. Therefore, we measured data while making a small amount of exhaust gas circulate to protect the burner. In this experiment, the NOx reached the highest value in the 70-80% rate of oxygen enrichment. Because of flue gas recirculation, the value of measured NOx concentration is smaller than that of the theoretical value. However, the characteristic of the theoretical value NOx appears clearly from acquired data.

It is understood that the oxygen enrichment strongly influences the generation of the NOx. considering the one whose NOx was 70ppm in air combustion becoming 700ppm in the oxygen enrichment 80%. (Figure.16) For the metal heating furnace whose maximum temperature is 1200°C, it is necessary to adjust the value of the NOx to at least 200ppm or less by the air pollution control law of Japan. Therefore, it is difficult to introduce simple oxygen-enriched combustion into the industrial furnace field. It is necessary to consider a low NOx measures to a great increase of the NOx at oxygen-enriched combustion.

![Figure.16. NOx Emissions with Oxygen-Enriched Combustion](image-url)
4.4. NOx reduction by FGR

In the experiment equipment, we have tried the reduction of the NOx due to the decrease in oxygen partial pressure in the combustion air by flue gas circulation.

Table 3 shows the data of the oxygen concentration in pseudo-air at oxygen-enriched combustion with flue gas recirculation.

<table>
<thead>
<tr>
<th>Oxygen-Enriched Rate</th>
<th>Gas (Pseudo-air)</th>
<th>Air (Pseudo-air)</th>
<th>O2 (Pseudo-air)</th>
<th>NOx</th>
</tr>
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<tbody>
<tr>
<td>% m3N/h m3N/h m3N/h m3N/h % ppm</td>
<td></td>
<td></td>
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<tr>
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<td>19.0</td>
<td>56.3</td>
<td>191.0</td>
</tr>
</tbody>
</table>

* FGR : Flue Gas Recirculation

NOx increases along with the rise of the temperature of the combustion air. Here, the recirculated flue gas was cooled to make the condition constant, and pseudo-air was supplied to the burner at about 100°C.

For 80% oxygen enrichment rate, the oxygen concentration in pseudo-air was 47% when the rate of flue gas recirculation was 2.0 m3N/m3N-fuel, and the actual value of the NOx was about 700 ppm. On the other hand, the oxygen concentration in pseudo-air was 22% when the rate of flue gas circulation was 8.0 m3N/m3N-fuel, and the value of the NOx was 76 ppm. As these results show, the effect of the NOx reduction of the exhaust gas recirculation is very large. (Figure 17)

In case of using waste heat from flue gas, it is available to use heat exchanger. However, there is few case of using waste heat recovery system such as waste heat boiler in industrial furnaces. It is desirable that to make good use of the heat within system.

Moreover, we tried to combustion test on the condition of not cooling the recirculated flue gas. Viewpoint of energy conservation, it is better to use recirculated flue gas that is not pass the heat exchanger. Figure 18 shows the result of experiment.
When exhaust gas is not cooled, the temperature of the pseudo-air supplied to the burner exceeds 350°C. It was able to be confirmed even here that the NOx was greatly reduced by recirculation.

4.5. NOx reduction by FDI (Fuel gas Direct Injection)

We tried to experiment FDI combustion to realize less amount of NOx using air fuel burner with recirculated flue gas. It is uncontrollable of the NOx formation when flue gas is not recirculated adequately in case that its temperature is too high to recirculate. Therefore, low NOx is realized by using FDI combustion technology. The recirculation of the furnace atmosphere is promoted by fuel gas jetting into the furnace. Gas nozzles are set by both sides of the burner as shown in Figure.10.

The effect of the NOx reduction with FDI combustion is shown in Table.4. The reduction rate of NOx is controlled by changing the setting angle of nozzle and PCD. NOx was decreased from 395ppm to 83ppm in case 1. In case 2, NOx was decreased from 1450ppm to 155ppm. It is realized that a low NOx combustion were possible even the rate of FGR is low.

We have to decide whether to use FGR or FDI combustion technology, considering oxygen enriched rate, the temperature of the furnace and NOx restriction.

Table.4. Actual Value of NOx Emission (FGR vs FGR+FDI)

<table>
<thead>
<tr>
<th>Case*1</th>
<th>NOx Reduction Method</th>
<th>Oxygen-Enriched Rate</th>
<th>FGR *2 [m3N/m3N-fuel]</th>
<th>O2 (Pseudo-Air)</th>
<th>O2 (Exhaust Gas)</th>
<th>NOx ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>395</td>
</tr>
<tr>
<td>1</td>
<td>FGR</td>
<td>40</td>
<td>47.3</td>
<td>31.8</td>
<td>8.5</td>
<td>395</td>
</tr>
<tr>
<td></td>
<td>FGR+FDI</td>
<td>40</td>
<td>46.1</td>
<td>31.6</td>
<td>7.9</td>
<td>83</td>
</tr>
<tr>
<td>2</td>
<td>FGR</td>
<td>60</td>
<td>37.6</td>
<td>44.1</td>
<td>10.0</td>
<td>1450</td>
</tr>
<tr>
<td></td>
<td>FGR+FDI</td>
<td>60</td>
<td>40.6</td>
<td>43.0</td>
<td>10.0</td>
<td>155</td>
</tr>
</tbody>
</table>

*1 Case1 : Furnace temp. 1200°C
Case2 : Furnace temp. 1300°C
*2 FGR : Flue Gas Recirculation

When exhaust gas is not cooled, the temperature of the pseudo-air supplied to the burner exceeds 350°C. It was able to be confirmed even here that the NOx was greatly reduced by recirculation.

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We have to decide whether to use FGR or FDI combustion technology, considering oxygen enriched rate, the temperature of the furnace and NOx restriction.
5 Summary

1. Success in applying oxygen combustion to convectional burner with solving NOx problem

We have succeeded in applying oxygen-enriched and oxy-fuel combustion system using recirculated flue gas to air combustion burner on the experimental furnace. The decrease of heat transfer by convection because of less volume of flue gas by oxygen combustions was feared in existing industrial furnaces or boilers. However, our proposal system enables to keep effectiveness of convectional heat transfer by combining this system with the oxygen combustion technology that adjusting the flue gas to the same amount as the air combustion. In order to enhance combustion efficiency, oxygen-enriched combustion is used by increasing the oxygen ratio in the oxidizer. However, since the flame temperature increases, NOx formation in the furnace seriously increases for low oxygen enrichment ratio. We mixed flue gas, air, and oxygen by using the flue gas recirculation system and generated pseudo-air of almost the same concentration of oxygen as air. The generation of NOx is remarkable in the state from 30 to 80% in the concentration of oxygen in air. However, to recirculate flue gas and decrease the concentration of oxygen in pseudo-air by mixing into the combustion air, it is possible to make NOx formation controlled. In addition, lowering the concentration of O2 in the pseudo-air down to 20.9%, it will be able to decrease the amount of NOx as same as that of air fuel combustion. And we found that less than 30% of CO2 concentration will control the NOx formation in practice.

It is accomplished that the concentration of CO2 reached over 90% in oxy-fuel combustion, and the amount of NOx is very few through the experiment. To apply this system to industrial furnace, it will be possible to reduce CO2 emissions by improvement of efficiency and realize zero-emission by CO2 recovery. And we believe it will lead to effective use of CO2 capture and sequestration. This system is applicable to other combustion instruments such as boilers.

Figure.18 shows idealized industrial furnace applying our system.

2. Feasibility of this system and oxygen supply

When the system that we propose is introduced into the market, there is problem that it costs producing or buying oxygen. Figure.19 shows the relation between the fuel reduction rate and the ratio of oxygen cost over fuel price. The lower the price of oxygen is compared with the price of the fuel, the more we can have huge advantage of the oxygen combustion. Moreover, the higher the temperature of the exhaust gas from furnace, the more it influences cost saving rate.

IEA (International Energy Agency) reported that oil prices will rebound to more than $100 a barrel as soon as the world economy recovers, and will exceed $200 by 2030. Put another way, current global trends in energy supply and consumption are patently unsustainable and it cannot avoid soaring of the
fossil fuel price in a mid/long-term viewpoint. When the price of oxygen relatively falls, the oxygen combustion technology will demonstrate great performance in both economically and efficiently.

### Figure 19. Relation between Saving Cost and O2-Cost / Fuel-Cost Ratio

3. Possibility of CO2 capture, sequestration and effective use

In the understanding of the characteristics of the oxy-fuel combustion, we want to refer the possibility of the CO2 capture and sequestration. In actual combustion, it is the above-mentioned that the majority elements of the flue gas are CO2 and H2O though some surplus oxygen is to be needed. One of the key technologies considered for carbon capture is the oxy-fuel combustion with recirculated flue gas that has been strongly considered for power plant retrofit and newly built plant in power generation industry. It can be said that this is an application of a technology concerned to recover CO2. It will be a big advantage to capture and sequestrate CO2 simply and efficiently in the power generating plants that consume a large amount of fossil fuels such as coal.

On the other hand, though the gross amount of energy consumption in industrial furnaces and boilers, the viewpoint of CO2 recovery was not fit. Because there were large numbers of industrial furnaces and boilers existing in various place. It can be said that it is common sense in this field to attempt the CO2 reduction by energy conservation up to the present.

The oxygen combustion technology has been remarked not only the technology of CO2 emission reduction but also the technology to realize CO2 recovery. Now, it is realized that applying oxygen combustion to common industrial furnaces by the system we proposed. And this system that can recover CO2 in high concentration on site will demonstrate great performance in case of using CO2 as inert gas or raw material in industrial processes.

Great amount of CO2 reduction or sequestration will be demanded in industrial furnace and boilers in the near future. It is no exaggeration to say that the application of the oxy-fuel combustion technology in industrial furnaces or boilers might have potential responding to such needs.

### References