Review

Development of oxy-fuel combustion technologies in TNSC

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Our oxy-combustion technology began with the introduction of oxy-fuel burner from the United States in 1970, and its purpose has changed significantly with the changing times. Initially, the purpose was to increase production by using it for industrial furnaces, then to save energy and environmental measures, and in recent years it has changed to contribute to carbon neutrality. Under these circumstances, this paper introduces the major oxygen combustion technologies that we have been developed over 50 years, and describes how they should be developed based on these technologies in the future.

1. Introduction

Use of oxygen for industrial purposes started with welding and cutting of steel. Industries in Japan earnestly began to adopt oxy-fuel burners at the early 1970s. It was at the end stage of the high-growth era, and as typified by auxiliary burners for electric arc furnaces for melting and recycling steel scrap, the purpose of using oxy-fuel burners was to increase productivity and contribute to the enhancement of production by simultaneously using oxy-fuel burners for conventional industrial furnaces.

However, since the first and second oil crises, industries in Japan experienced the low-growth era. At the same time, oil prices skyrocketed, and the purpose of using oxy-fuel burners changed from increasing production to reducing energy and costs.

In the 1990s, as the worldwide consumption of fossil fuels increased, problems arose, such as environmental issues concerning global warming resulting from CO2 emissions and acid rain caused by nitrogen oxides as well as recycling issues with the aim of effectively using the earth's limited resources. Accordingly, the purpose of oxy-fuel combustion technology was redirected to the contribution to solving environmental issues in addition to energy saving[1). Furthermore, as the result of the Kyoto Protocol in 1997 and the Paris Agreement in 2015, reduction of greenhouse gases became an important issue, and environmental issues began to greatly influence the industrial structure so as to further increase energy efficiency, convert the use of fossil fuels to renewable energy, and so forth.

Under these circumstances, TNSC has been promoting technological development based on the accurate understanding of customer needs. In this paper, we will introduce characteristics of oxy-fuel combustion and oxy-fuel combustion technologies that we have developed over the past 50 years, and we will offer an outlook for the future based on the recent market trends as well as environmental issues.

2. Characteristics and effects of oxy-fuel combustion

2.1 Flame temperature and burnt gas composition

The characteristics of oxy-fuel combustion are described in comparison with air-fuel combustion. Figure 1 shows the relationship between the oxygen concentration in oxidant gas when methane is used as a fuel and the equilibrium flame temperature (adiabatic flame temperature) in the adiabatic state. Figure 2 shows the relationship between the oxygen concentration in the oxidant gas and the equilibrium gas composition, specifically, concentration of radical substances. Those figures indicate that flame temperature of oxy-fuel combustion is approximately 800 K higher than that of the air-fuel combustion, and a large quantity of radical substances are included in the high-temperature flame.

When methane is burned, chemical reactions in air-fuel combustion and oxy-fuel combustion are expressed by Eq.(1) and Eq.(2). That is, Eq. (1) and Eq. (2) indicate that oxy-fuel combustion which does not contain nitrogen as an inert gas in the right-hand side showing an exothermic reaction is energetically advantageous.

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Air-fuel combustion	$CH_4 + 2O_2 + 8N_2 \rightarrow CO_2 + 2H_2O + 8N_2 + Q$	(1)
Oxy-fuel combustion	$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O + Q$	(2)

There are two types of heat transfer from the flame to an object to be heated: convective heat transfer which is direct heat transfer and radiative heat transfer which is indirect heat transfer. They can be expressed by Eq. (3) and Eq. (4), respectively.

Radiative heat transfer	$q = \varepsilon\sigma\left(T_f^4 - T_s^4\right)$	(3)
Convective heat transfer	$q = h\left(T_f - T_s\right)$	(4)

q : Heat flux [W/m^2]
T_f : Flame temperature [K]
T_s : Temperature of object to be heated [K]
h : Heat transfer coefficient [W/m^{2}K]
ε : Emissivity [-]
σ : Stephan-Boltzmann constant [W/m^2 K^4]

As mentioned above Compared with air-fuel combustion, flame temperature of oxy-fuel combustion is approximately 800 K higher. Eq. (3) and Eq. (4) indicate that this characteristic significantly contributes to the increase in efficiency of forced convective heat transfer and radiative heat transfer.

Also, as shown in Eq. (1) and Eq. (2), combustion flame of CH_4 is a mixture of CO_2 and H_2O. The mixture of CO_2 and H_2O dissociates into carbon monoxide (CO), hydrogen (H_2), and oxygen (O_2) at temperatures between 1800 and 2300 K; and at higher temperatures, it dissociates into oxygen atoms (O), hydrogen atoms (H), and hydroxyl ions (OH). Since flame temperature of the oxy-fuel burner exceeds 2300 K, the thermal dissociation occurs. When this flame collides with objects to be heated, such as ceramics particles, etc., flame temperature decreases, generating dissociation products of CO_2 and H_2O, thereby releasing latent heat of dissociation. As a result, the heat transfer efficiency is further increased [2].

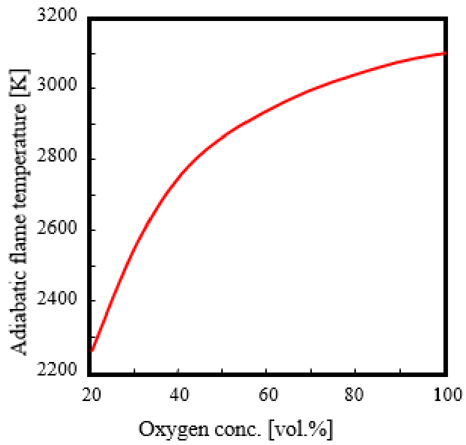


Fig. 1 Relationship between oxygen concentration and theoretical flame temperature

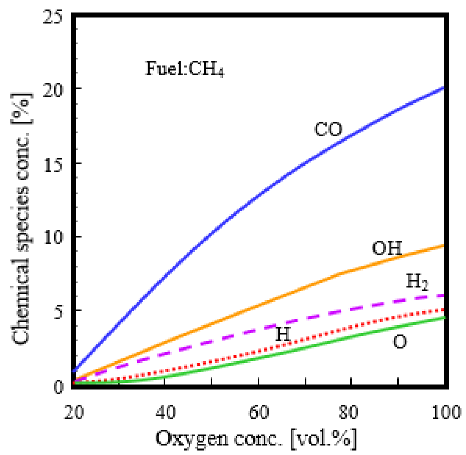


Fig. 2 Relationship between oxygen concentration and chemical species concentration in burnt gas

2.2 Burning velocity

Figure 3 shows the relationship between the oxygen concentration in the oxidant gas and the burning velocity of typical gas fuels. Compared with air-fuel combustion, burning velocity of oxy-fuel combustion is three to six times higher, and a high-speed flame can be formed stably. By making high-speed flame collide with objects to be heated and melted at high temperatures and with the addition of high forced convective heat transfer and latent heat resulting from the recombination with the above-mentioned dissociation products, it is possible to achieve highly-efficient heat transfer. It is also possible to burn fuels of relatively low burning velocity, for example, ammonia, in a stable manner. Furthermore, stable combustion can be maintained even when the amount of oxidant is increased or decreased with respect to the stoichiometric ratio required for fuel combustion. Therefore, the combustion atmosphere can be controlled to an oxidation or reduction atmosphere, and combustion characteristics suitable for various applications are provided.

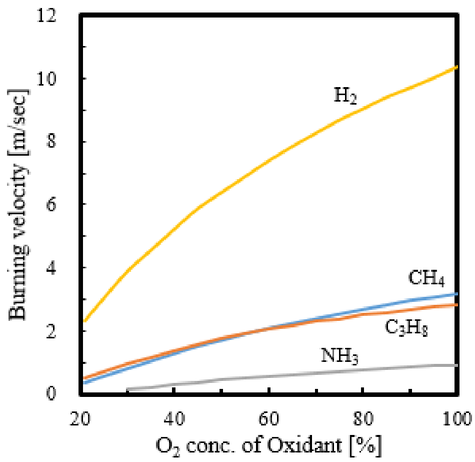


Fig. 3 Relationship between oxygen concentration in combustible gas and burning velocity

2.3 Thermal efficiency of oxy-fuel combustion

In oxy-fuel combustion, the amount of flue gas is approximately one forth because nitrogen is not contained in the oxidant gas. Therefore, the sensible heat of the flue gas is greatly reduced, and significant energy saving is possible. As an example, Fig. 4 shows the relationship between the flue gas temperature and the thermal efficiency where the thermal efficiency in methane combustion is defined as (calorific value of methane - sensible heat of flue gas) / (calorific value of methane). In this case, thermal efficiency indicates the proportion of heat quantity used in the combustion field (combustion furnace, etc.), and it is used to transfer heat to the objects to be heated and melted as well as heating the furnace body.

According to Fig. 4, for example, when flue gas temperature is 1700 K, thermal efficiency is approximately 40% in air-fuel combustion and as high as approximately 80% in oxy-fuel combustion. Since efficiency becomes twice as much in this case, energy consumption becomes half. As a result, it is possible to significantly save energy by means of oxy-fuel combustion.

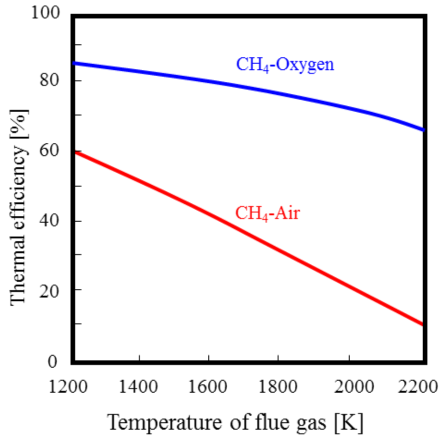


Fig. 4 Relationship between flue gas temperature and thermal efficiency in a furnace

2.4 Analysis of oxy-fuel combustion

Combustion occurs as the result of chemical reactions of chemical species in the fuel and is greatly affected by the method of feeding fuel and oxidant gas into the combustion field in addition to the burning velocity of fuel. That is, the flame temperature, gas velocity, and radiation intensity change depending on the burner nozzle structure, and it is necessary to provide the flame characteristics suitable for utilization fields. For the feedback to the burner nozzle design, it is important to understand flame characteristics, and therefore, measurement and analysis should be performed.

Our oxy-fuel combustion technologies have been developed by responding to liquid fuels, such as kerosene and heavy oil, gaseous fuels, such as natural gas and propane, and solid fuels, such as pulverized coal and char. In addition, because temperature of the reaction field of oxy-fuel combustion becomes extremely high, existing measurement methods cannot be directly used in many cases. Thus, we proceeded with development by selecting appropriate measurement methods suitable for the combustion atmosphere of utilization fields.

(1) Measurement of combustion

Atmospheric temperature is generally measured directly by a thermocouple. However, in flame temperature measurement, there are problems that accurate gas temperature cannot be measured due to the inflow and outflow of radiant heat to the thermocouple element, and that high temperature flames such as oxy-fuel combustion flames exceed the measurable range of thermocouples. Therefore, the suction pyrometer was improved and changed to a water-cooled type, which was used for measurement. The structure is shown in Fig. 5. The suction pyrometer utilizes a method of measuring temperature by a thermocouple while sucking an atmospheric gas. This method removes the effects of radiation and reduces the gas temperature to a measurable range with a thermocouple by water cooling. However, in this situation, gas temperature is not accurate; therefore, decrease in gas temperature by means of the water-cooled system is separately corrected by a measurement instrument using an optical method.

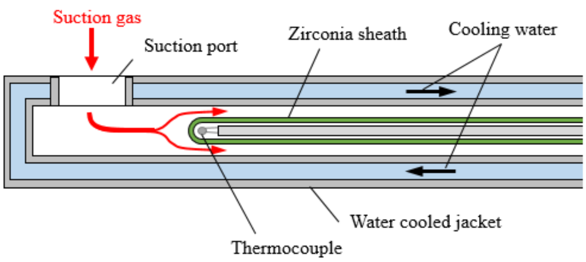


Fig.5 Schematic of the water-cooled suction pyrometer tip

There are roughly two types of measurement methods of attaining more detailed flame temperature distribution. One type of methods includes Rayleigh scattering, Raman scattering, CARS, LIF, etc. which utilize the laser to measure a number of local values so as to attain distribution. The other type of methods combines a radiation method, sonic method, or holography method with the computed tomography (CT) technique to attain distribution from projection data. From those methods, we selected the radiation method whose measurement principles are relatively simple. The radiation method includes two techniques: one uses radiation or absorption of H_2O or CO_2 gas, and the other uses radiation of solid like soot particles. We have developed a method to measure a two-dimensional distribution of flame temperatures by combining the radiation method with the CT technique.

In the measurement of gas velocity in the flame, velocity of each portion of the flame was measured by directly inserting a water-cooled pitot tube into the flame regardless of types of fuel to be burned [2].

Since fuel spray is important for the combustion of liquid fuel, the diameter and dispersion of droplets due to fuel spray have a great influence on the combustion situation, and these are important parameters for the design of the burner nozzle. For this reason, we measured the fuel spray state by means of an optical technique. As spray measurement methods, a high-speed projection method, immersion method, laser diffractometry, hologram method, etc. have been conventionally used. However, those methods have a limitation in capturing local spraying behaviors and fluctuations of spray time, and we used the Phase Doppler Anemometry (PDA) using the laser [3, 4].

The PDA technique has two advantages as described below.

(1) It does not disturb the field because of non-contact measurement.

(2) It is capable of measuring high spatial and temporal resolution.

The PDA technique adopts the principles of the Laser Doppler Velocimeter (LDV). When particles floating in a fluid are irradiated with laser light, frequency of the light scattered from the particles changes by the Doppler effect according to the moving speed of the particles. This technique obtains the moving speed of particles by detecting the changes in frequency.

To develop a burner that can achieve an optimal combustion method, it is very important to understand fuel spray properties.

Furthermore, in order to evaluate specific flame combustion characteristics necessary for assuming use situations, such as industrial furnaces, etc., we measured the radiative heat flux and the convective heat transfer.

With use of a 2π furnace radiometer to measure the radiative heat flux, we measured the distribution of radiative heat flux from the flame during the exposure to the atmosphere or combustion in the furnace and evaluated the validity of burner design as well as adaptability to use situations. To measure the convective heat transfer, we fabricated a water-cooled board, shown in Fig. 6, by combining Cu tubes. A method of measuring the amount of heat transfer was used from the change in water temperature in and out when a flame collided with this water cooling plate [5].

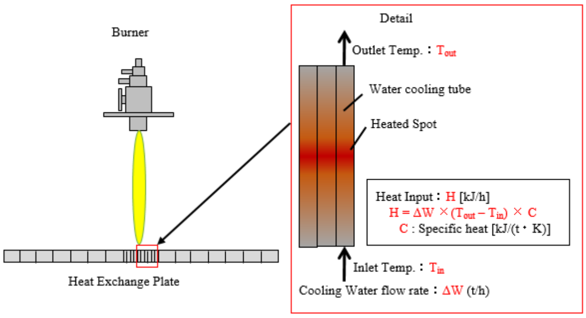


Fig.6 Experimental equipment for measuring heat transfer distribution

(2) Analysis of combustion

We proceeded with the development of the burner by repeatedly performing design, production, and combustion testing so as to attain a desired combustion state. To adapt the burner to industrial furnaces, we followed the procedures of preliminary laboratory testing and testing using actual furnaces. Since testing using a currently-operated actual furnace involves taking risks, we utilized numerical simulation for the purpose of accelerating development, smoothly applying the burner to actual furnaces, and providing feedback to the furnace design.

Burners have been used in a variety of fields, such as glass melting furnaces, metal heating furnaces, and also as single-unit burners. As a typical example, Fig. 7 shows an example that simulates the distribution of temperatures in a furnace when oxy-fuel combustion is applied to the glass melting furnace [6]. We uniquely developed this analysis code because no general-purpose code was available for oxy-fuel combustion at the time of development. We currently use several general-purpose codes to develop applications.

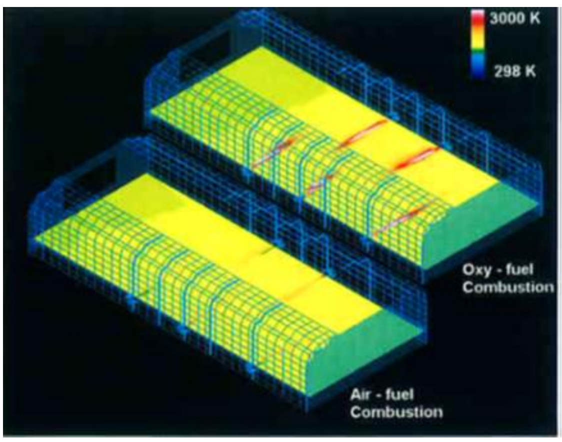


Fig. 7 Temperature distributions in the glass furnace

3. Endeavor to develop oxy-fuel combustion technologies (history)

TNSC introduced oxy-fuel burner technology from U.S. Air Reduction Company in 1970. This oxy-fuel burner adopted a system that ejects fuel heavy oil and oxygen from the nozzle tip so as to perform diffusive mixing combustion. The oxy-fuel burner was mounted to a melting furnace or a sintering furnace together with an air burner so as to increase productivity of the furnace. The oxy-fuel burners were mostly used for melting aluminum in the United States, however, in Japan, they were mainly used for electric arc furnaces for steel scrap to save energy. They were also used for aluminum melting furnaces, heat-resistant cement melting furnaces, and alumina sintering kilns. However, as use purposes diversified as times changed, we have devoted ourselves to develop our own unique products since 1975.

In 1976, we participated in the joint project of developing a combustion decomposition technology for polychlorinated biphenyl (PCB) with the National Institute of Industrial Safety of the Labor Ministry, and we successfully achieved a decomposition rate of 99.99999% by using PCB itself as a fuel instead of using an auxiliary fuel [7]. Since this burner was able to generate high-temperature, high-speed flame with use of general fuel like heavy oil etc., we manufactured the burner on a commercial basis as an oxy-fuel burner "Super OFB-L type". This became the starting point for our oxy-fuel combustion technology and set the groundwork for the technological development over the following 50 years as shown in Fig. 8 (following page).

Figure 9 shows the application of each technology. TNSC has applied the technologies mainly to the metal field, and then to the glass, energy, and waste fields.

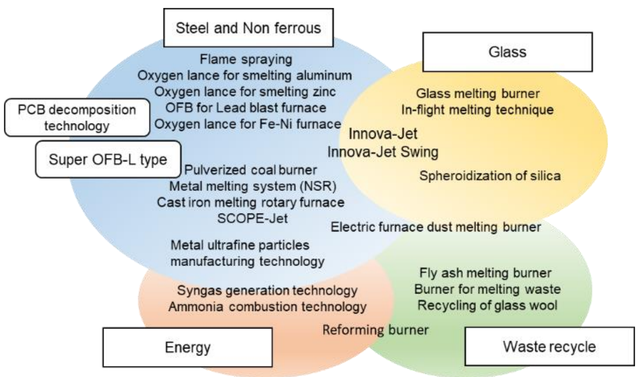


Fig. 9 Oxy-combustion technology initiatives

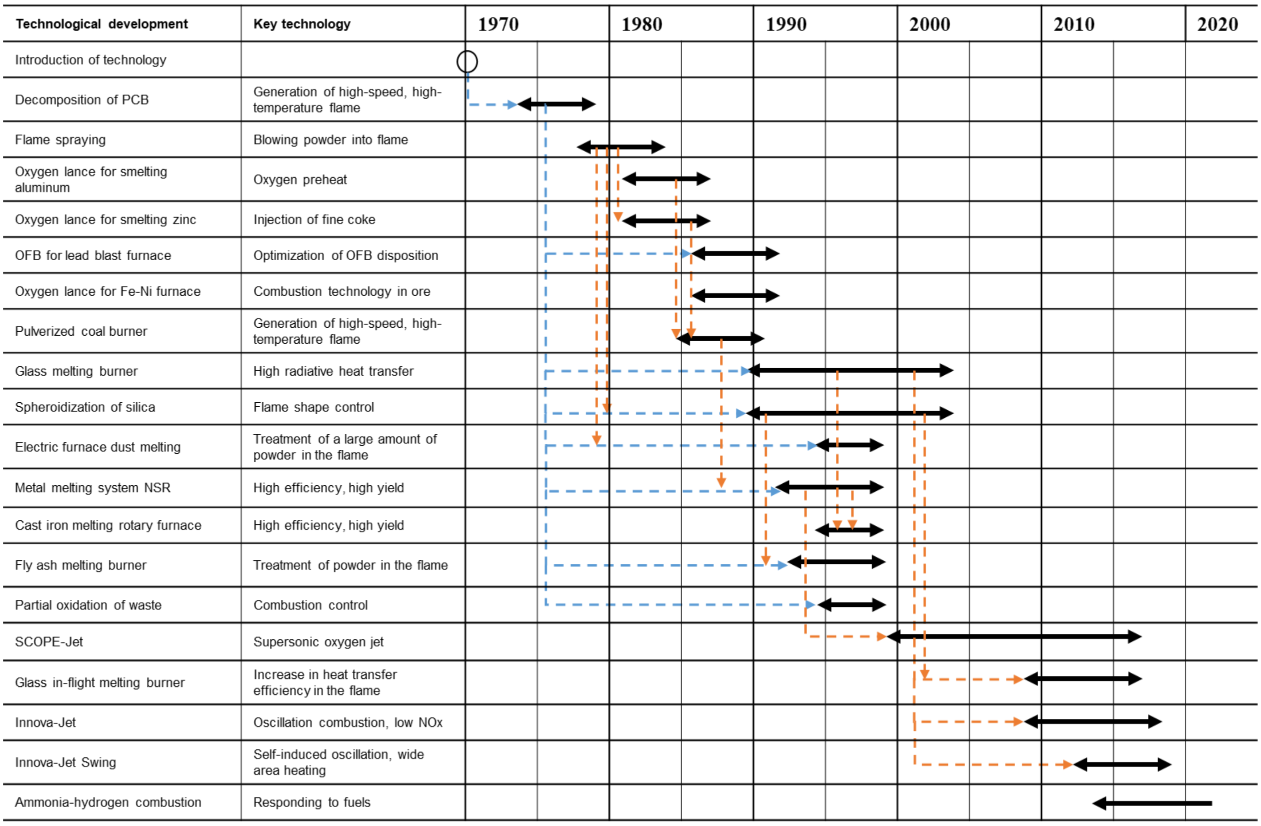


Fig. 8 History of oxy-combustion technologies developed by TNSC

In the 1980s, TNSC started to develop a variety of applied technologies utilizing oxy-fuel combustion. Specical.¸, we proactively undertook new projects in the metal field. The "flame spraying" is a technology developed for the purpose of repairing a furnace wall of the converter furnace by means of hot processing. Refractory powder is injected into the propane-oxygen flame, melted in the flame, and coated onto the damaged part of the furnace at high speed. Compared with conventional wet type methods, it has been confirmed that the repair layer generated by this technique is 5 to 7 times durable [8].

The "oxygen lance for smelting aluminum" is based on the technology to blow 1073 K to 1273 K high-temperature oxygen through the tuyere into a blast furnace that directly reduce alumina to produce aluminum. At the development phase, we established a basic technology of the high-temperature oxygen generation process and a technology of the water-cooled jacket that can withstand an ultra-high-temperature atmosphere [9, 10]. The "oxygen lance for smelting zinc" is an injection lance (oxygen jet: Mach number 1 to 1.5) that can blow raw material ore and coke used for melting reduction on high-speed oxygen jet and blows them into the molten zone in the furnace. This technology was the starting point of the development of supersonic oxy-fuel combustion technologies. The "lead blast furnace OFB" is an applied technology to reduce the unit consumption of coke in the lead blast furnace by installing an oxy-fuel burner at the tuyere of the blast furnace and also melting an unfused part at the bottom of the furnace. The "Fe-Ni oxygen lance" is based on the technology to use an oxy-fuel burner for thermal compensation in the electrical furnace for melting and reducing nickel-containing ore. Furthermore, we developed a technology to promote efficient combustion by inserting an oxy-fuel burner approximately 2 meters into high-temperature ore and a technology used to prevent wear and water leakage of the water-cooled jacket of the burner caused by the flow of high-temperature granular ore [11].

Around the same time, alternative energy to oil began to be discussed after oil crises, and we started to develop a technology to burn coal that is richly deposited. We developed a "pulverized coal burner using high-temperature oxygen" having the same combustion characteristics as a heavy oil-oxygen burner [12]. Successively, as one of its applied technologies, we developed a "metal melting system (NSR)" to melt steel scrap by use of only oxy-fuel burners [13]. Through development of this technology, we learned an iron scrap melting technology using the oxy-fuel burner and a basic technology of steelmaking. We then utilized those technologies to the applications in the steel market.

In the steel field, we developed "high-speed oxygen burner lance: SCOPE-JET" for the purpose of improving efficiency and reducing power consumption for an arc furnace (EAF) to melt steel scrap. We also developed "Innova-Jet" intended for high-efficiency preheating for the heating furnace, ladle furnace, etc. We brought those products to the market. These technologies will be described later in this paper.

From 1990s, we gradually shifted the course of technological development from the development of single-unit oxy-fuel burners to "applications" specialized in the market. Specifically, the worldwide enhancement of consciousness about the reduction of CO_2 emissions resulting from the enforcement of the Kyoto Protocol and decrease in oxygen price due to the appearance of the PSA oxygen generators helped the application of oxy-fuel combustion to expand to the glass and ceramics fields in addition to the metal field in which oxy-fuel combustion had been widely used. In the metal field which had been a target market, convective heat transfer type oxy-fuel burners with high flame gas speed were mainly used, and we had developed burners for the purpose of promoting mixture of fuel and oxygen and increasing burning velocity to increase flame temperature. However, in the glass melting furnace, radiative heat transfer from the burner is important. Therefore, we focused on the amount of soot generated in the flame and its residence time as for a way to increase radiative heat transfer and developed a "glass melting burner" in which the liquid fuel atomization method and the oxygen blowing speed had been optimized. By applying oxy-fuel combustion to the glass melting furnace based on this technology, we have successfully achieved energy saving, low NOx, and high quality [6, 14].

The above-mentioned "flame spraying" is a technology to heat and melt ceramic particles in the high-temperature flame, which served as a basic technology for the "silica spheroidization system: CERAMELT" described later. The CERAMELT is made up of a series of systems from supply of powder, melting, and to collection processes. This technology was expanded to be used for melting electronic materials, such as alumina, ferrite, powdered glass, etc., in addition to silica [15-18]. Other than that, as a technology to treat powder in the flame, this technology was applied to the "fly ash melting burner [19]" which treats fly ash from incinerators, the "oxy-fuel burner for melting and solidifying waste [20]" which melts dust generated in the electric furnace, and the "glass wool recycling system [21]" which melts and recycles glass wool used for adiabatic material. Those burners and systems have achieved satisfactory results in the industrial waste recycle market.

Furthermore, in recent years, also in the above-mentioned glass industry, an innovative glass manufacturing technology "in-flight melting technique" has been developed [22]. As one of important element technologies for this development, the oxy-fuel burner technology was adopted in which glass raw material powder is loaded in the flame and efficiently heated and melted. Thus, potential of powder treatment technologies using oxygen-burning flame has been expanding [23].

In the waste field, the gasification melting furnace was put into practical use to recover energy. By making use of the characteristics that increase fuel burning velocity in oxygen and facilitate the oxidation-reduction control, TNSC developed a "reforming burner" for generating hydrogen and carbon monoxide by partially oxidizing pyrolysis gas, pyrolysis oil, and char in the waste [24]. Furthermore, since high-temperature reduction atmosphere can be generated in the flame, by performing reduction treatment of metallic compounds in the flame, we expanded the technology to develop an "ultrafine metal particle manufacturing technology". As this technology makes it possible to change oxides, such as nickel, copper, etc., into ultrafine particles of 100 nm or less, it is expected to be widely used for multilayer ceramic capacitors and conductive paste [25-27]. Furthermore, based on the combustion control technologies, we developed a technology to obtain syngas by partially oxidizing natural gas [28].

As stated above, TNSC accurately understands the needs from various fields, performs feedback of those needs to the technological development, and is striving to deepen the oxy-fuel combustion technology to respond to the market trends.

4. Major oxy-fuel combustion technologies

History of the development of oxy-fuel combustion technologies was overviewed in the previous section. In this section, key technologies will be described in detail.

4.1 Pulverized coal burner using high-temperature oxygen

Conventionally, liquid fuel, such as kerosene, heavy oil, etc., and gas fuel, such as propane, hydrogen, etc., had been used for oxy-fuel combustion. Since the occurrence of oil crises, coal was reconsidered from the aspect of alternative energy to oil. However, coal is solid and burning velocity is lower than liquid fuel and gas fuel, and it was not suitable for use purposes requiring high-temperature flame, for example, metal melting of steel scrap, etc.

We thought it possible to improve the disadvantage of combustion performance by use of preheated high-temperature oxygen to increase burning velocity and generate high-temperature flame. We then started to develop a high-temperature oxygen generation process, select metal and ceramic material used with high-temperature oxygen, perform durability testing, examine basic combustion characteristics of pulverized coal [29-31], and proceeded with the development of the pulverized coal oxy-fuel burner [12].

Figure 10 shows the effect of preheated oxygen temperature on the combustion rate and the effect on flame temperature is shown in Fig. 11 at a pulverized coal combustion scale of 150 kg/h. Figure 10 indicates that the combustion rate increases as the preheated oxygen temperature increases, and at the oxygen preheating temperature of 1073K, the combustion rate is 0.75 at a distance of 0.5 m from the burner tip and 0.97 at a distance of 1.0 m. As shown in Fig. 11, it was found that a high-temperature flame of 2900 K or higher can be obtained by preheating oxygen to a flame temperature is 2500 K when oxygen at room temperature is used.

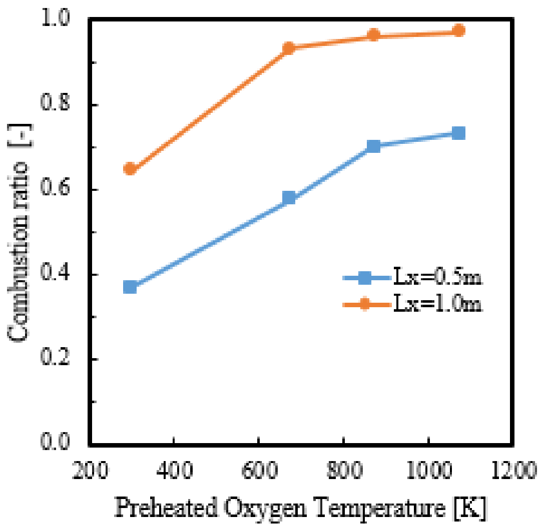


Fig.10 Effect of oxygen preheated temperature on combustion rate

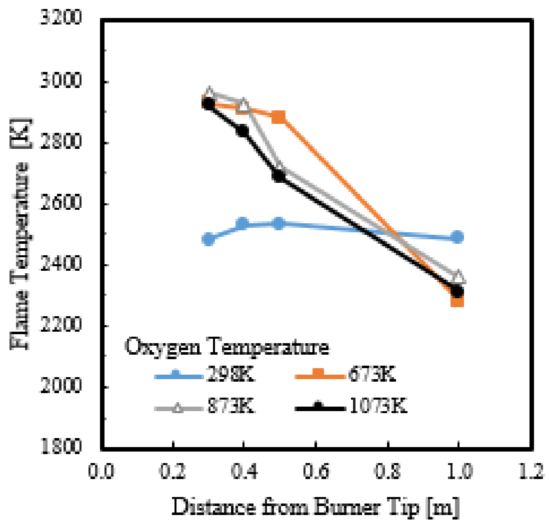


Fig.11 Effect of oxygen preheated temperature on flame temperature

Upgrading the development scale based on these results, we developed a high-performance pulverized coal burner (pulverized coal 500 kg/h, oxygen 750 Nm^3/h). Required preheated oxygen temperatures vary depending on the combustion scale of pulverized coal, ranging between 570 k and 1070 K.

This technology was highly evaluated in relevant industries and academic societies as a technology to burn pulverized coal safely and efficiently using high-temperature oxygen. This technology became an underlying technology for metal melting technologies described in the following section. As for oxygen preheating methods, we developed an indirect heating method by means of heat exchange and a method of directly heating oxygen by burning fuel in oxygen. We thus established the basic technology of preheating oxygen [12].

4.2 Metal melting system "NSR"

In Japan, approximately 30% of crude steel production results from recycling steel scrap. Most of it is melted for recycling using electrical arc furnaces (EAF), consuming a huge amount of electricity.

Although electricity is high-quality, easy-to-use energy, when taking into account energy input in the generation of electricity, the effective thermal efficiency of steel scrap is reduced to as low as approximately 25%. Joining the project subsidized by the Ministry of International Trade and Industry (of those days), we established a basic technology to directly melt steel scrap for recycling only by the oxy-fuel combustion [32-33].

This is a process in which 50 to 60% of heat input into the oxy-fuel burner is effectively utilized, making it possible to significantly save energy.

Figure 12 shows the basic structure of the furnace in which steel scrap is melted for recycling only by the oxy-fuel burner. The furnace consists of a melting zone, metal pooling zone, and holding zone, and each zone is equipped with more than one oxy-fuel burner. Fuel can be selected from gas, liquid, and solid fuels according to the situation.

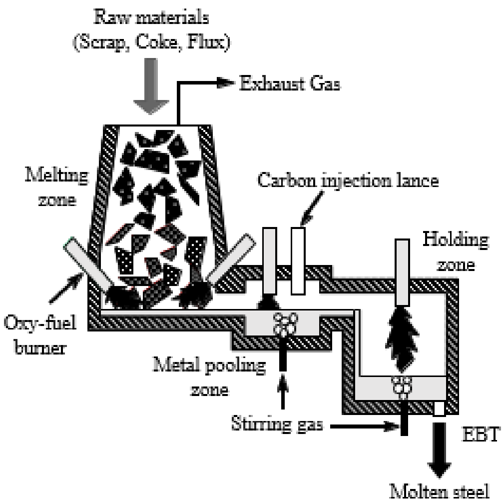


Fig.12 Schema of NSR process

Steel scrap fed from the furnace top of the melting zone along with sub-materials is continuously melted by more than one oxy-fuel burner installed at the bottom of the furnace and flowed to the metal pooling zone and then to the holding zone. In the holding zone, molten steel is temporarily stored, smelted, and exposed to increasing temperature, and then tapped from the bottom of the furnace at 1870 K or higher. All combustion flue gases generated in the metal pooling zone and the holding zone pass through the steel scrap layer in the melting zone, and therefore, temperature of flue gas coming from the furnace top of the melting zone is as low as 500 to 600 K. For the reason, a thermally efficient process has been achieved. With the melting capacity of 6 t/h of a pilot scale, we investigated thermal efficiency of this process, controllability of components, such as carbon (C) and phosphorus (P), and dust emission characteristics. As a result, when compared with general EAFs, we found that approximately 40% of energy saving had been achieved (Fig. 13).

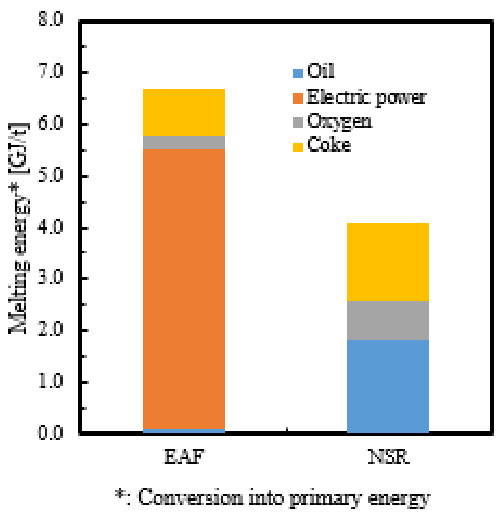


Fig.13 Comparison of melting energy

This newly developed process was a continuous type process. It was not suitable for the needs at that time, and there were still more development factors such as the durability of refractories. Therefore, this new technology was not put into practical use. However, we made use of the findings from this development for the applications to the electric furnace.

4.3 High-speed oxygen burner lance: SCOPE-JET

Based on the technologies cultivated through the development of the metal melting technology, we developed a burner lance SCOPE-JET, which generates supersonic oxygen jet, as part of energy saving technology to be used for electric arc furnaces for melting and recycling steel scrap [34-37]. The SCOPE-JET is a burner lance that functions as an oxy-fuel combustion burner and a lance for blowing oxygen into molten steel.

Figure 14 is a conceptual diagram of the supersonic oxygen jet. This lance features its simple nozzle structure. With this very simple nozzle structure, it is possible to generate stable flame around the supersonic oxygen jet and also significantly suppress the decrease in the velocity and oxygen concentration of the supersonic oxygen jet.

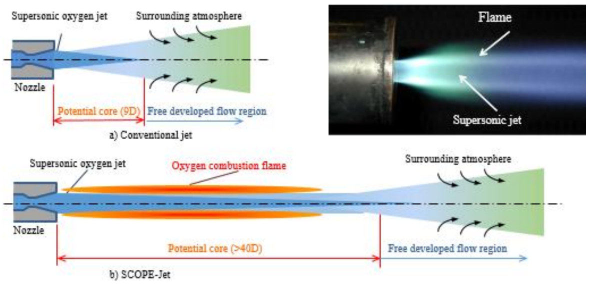


Fig.14 Schematic diagram of supersonic oxygen jet

Market deployment of the SCOPE-JET started in 2003 as an auxiliary melting technology for electric arc furnaces. As shown in Fig. 15, in the steelmaking process of the electric arc furnace, the SCOPE-JET functions as an oxy-fuel burner to promote scrap melting at the "melting period" immediately after steel scrap was fed, and then it functions as an oxygen lance at the "refining period" where scrap had melted and dropped. It is an advantage over conventional auxiliary burners that do not have an oxygen lance function. As a result, it is possible to greatly contribute to the increase in productivity of the steelmaking process in the electric furnace and the reduction of unit consumption of energy.

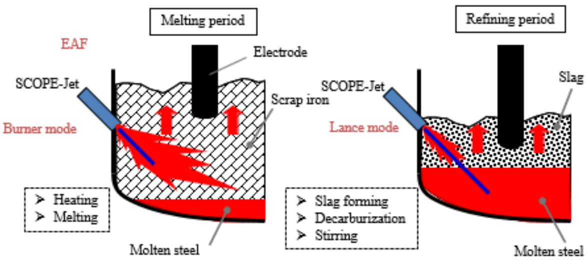


Fig.15 Schematic diagram of SCOPE-JET operation in electric arc furnace

The SCOPE-JET can utilize low-pressure, low-calorie fuels including Cokes Oven Gas (COG) in addition to gas fuels, such as natural gas and propane gas, and liquid fuels, such as kerosene and heavy oil.

Furthermore, at the development phase, the oxygen jet speed was Mach number 1.5, but performance has been improved to Mach number 2.0. Thus, the function as an oxygen lance at the refining period has been enhanced.

4.4 Ultra-low NOx oxygen-enriched burner: Innova-Jet

To apply oxygen-enriched combustion to high-temperature heating furnaces, one issue to be solved is to suppress nitrogen oxides (NOx) generated as temperature of the furnace increases. This problem was solved by an oxygen-enriched burner, Innova-Jet, that can significantly reduce NOx at the time of oxygen-enriched combustion [38-41]. This burner uses TNSC-original oscillation combustion in which the oxygen concentration and the oxygen ratio in an oxidant is periodically changed, and the burner is capable of reducing NOx emissions to one twentieth, compared with general oxygen-enriched combustion (Fig. 16). In addition, by greatly fluctuating flow rate of oxidant gas periodically, gas in the furnace is stirred, and thereby heat transfer efficiency is increased by approximately 10%. This technology was applied to preheating of the ladle furnace and achieved the fuel reduction of 30 to 40%.

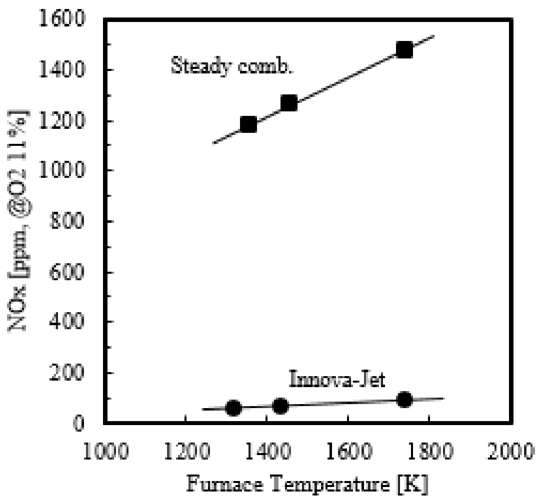


Fig.16 Comparison of NOx emission characteristics between conventional oxygen-enriched combustion and Innova-Jet

Thermal efficiency of oxy-fuel combustion and oxygen-enriched combustion can be improved by reducing the amount of flue gas. On the other hand, locally high temperature tends to occur due to high flame temperature, and therefore, a technology to uniformize temperature distribution is required. To solve this problem, we developed Innova-Jet Swing that utilizes the self-induced oscillation phenomenon [42-45]. The self-induced oscillation is a phenomenon wherein fluid independently oscillates without being operated externally, and this phenomenon can be produced by combining the "Coanda effect" by which fluid flows along the vicinity wall with a specialty nozzle structure that induces a force to pull the fluid away from the wall (Fig. 17). By applying this phenomenon to the burner nozzle design, it is possible to periodically change the direction of the flame, thereby increasing the heating area. As an example, Fig. 18 shows the situation in which the flame oscillates right and left. Furthermore, since this burner does not require a mechanical drive unit, the structure can be simple and maintenance is easy.

As an application example, when preheating the tundish* under 40% oxygen-enriched operating conditions, we confirmed that the amount of fuel used can be reduced by 40% compared with air-fuel combustion.

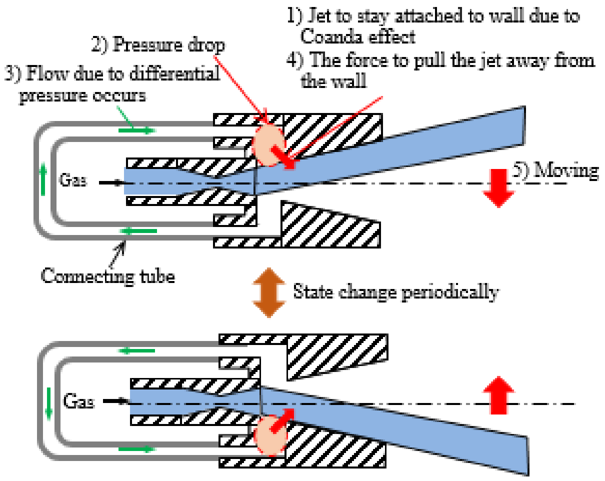


Fig.17 Schematic diagram of self-induced oscillation nozzle

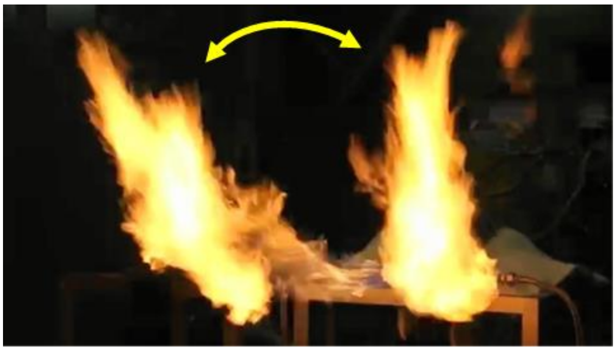


Fig.18 Combustion state of self- induced oscillation burner

*Tundish: A tray, provided in the continuous-casting machine, which temporarily receives molten steel poured from the ladle furnace to a casting mold so as to further remove inclusions

4.5 Silica spheroidization technology: CERAMELT

This is a technology to feed ground ceramics in the oxy-fuel combustion flame and melt them by applying the above-mentioned flame spraying technology [15].

As the result of downsizing and high-performance of semiconductors, integration of semiconductor devices has been increased. Associated with this trend, semiconductor encapsulating methods have been changed. Semiconductors are encapsulated for the purpose of protection from the external environment and addition of the strength. Epoxy resin which includes ground molten silica as a filler has been conventionally used as encapsulating material. However, as the integration level increases, spherical amorphous silica with increased flowability is mainly used with the intention of high-density packing.

To obtain spheroidized silica, highly-pure powdered natural quartz is injected into the high-temperature oxy-fuel combustion flame, completely melted in the flame, and vitrified to form spherical glass. The melting point of highly-pure silica is as high as 1923 K and the viscosity of the melt is high. Therefore, to completely melt, spheroidize, and vitrify highly-pure silica, sufficient residence time at high temperature is required. We developed a burner and a system that can produce the flame characteristics suitable for spheroidization and thus established a spheroidization technology. The system flow is shown in Fig. 19.

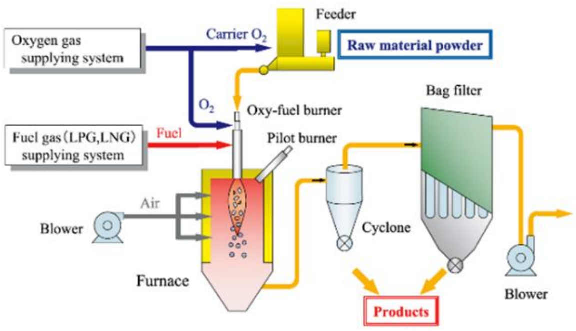
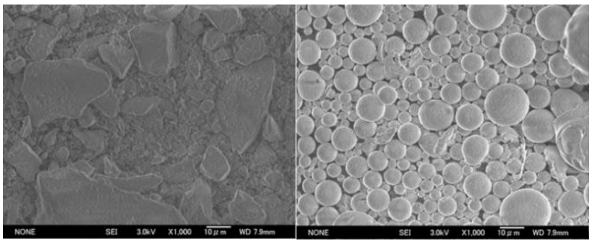


Fig.19 Schematic diagram of apparatus

Figure 20 shows the shapes of raw material silica and spheroidized silica. The raw material silica is crystalline, and the vitrification rate of the spheroidized silica after treatment is 97% or more, which makes it possible to obtain high-quality products.



(a) Raw material (b) products

Fig.20 Pictures of raw material and products

Recently, with the advancement of semiconductors, the spheroidization system needs to handle a variety of materials with different particle diameters [16-18], and efficient approach is necessary to accelerate development. Accordingly, we utilized numerical simulation and developed a technology to presume powder behaviors in the oxy-fuel combustion flame [46].

An example of the simulation is shown in Fig.21. By performing analysis of two kinds of raw material with different particle diameters (15 µm, 5 µm), we found the possibility that the difference in dispersibility resulting from kinetic momentum of particles may affect the heating performance of material with a small particle diameter. As the result of feedback of the finding to the burner design, it was consistent with the experiment result. Thus, we established a technology to presume flow of powder and temperature behaviors in the oxy-fuel combustion flame by use of numerical simulation.

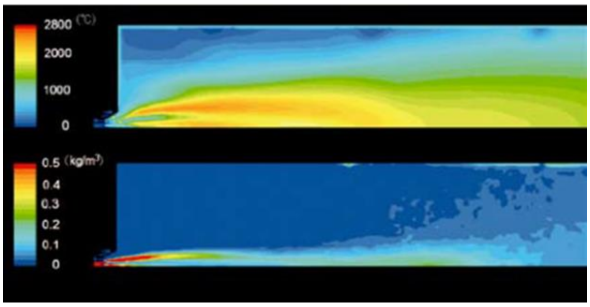


Fig.21 Contour of gas temperature and particle concentration

4.6 Oxy-fuel burner for the glass field

Generally, in the glass industry, glass raw material is melted by means of air-fuel combustion using a huge amount of fuel. Major environmental problems in this industry are to save energy and suppress NOx emissions resulting from combustion. In addition, since air-fuel combustion is not thermally efficient, large heat recovery equipment and regenerative furnaces are required. For the reconstruction, a huge quantity of bricks and industrial waste contaminated by boron, lead, coloring agents, and heavy metals contained in add-in materials is generated, and accordingly, reduction of those materials is another problem to be solved.

Europe and the United States took the initiative in substituting oxy-fuel combustion for air-fuel combustion mainly for the purpose of reducing NOx emissions. Because Western fuel is mostly natural gas, it was difficult for Japanese glass industry that mainly uses liquid fuel to directly introduce the Western technologies. For this reason, a domestic glass manufacturer that had been proactively coping with environmental issues focused on the alternative technology of the Western oxy-fuel combustion and launched a joint development project with TNSC in 1990. We had been engaged in the development of oxy-fuel burners for melting glass using heavy oil as a fuel and successfully put the new burners into practical use two years later.

Heat transfer in glass melting is radiative heat transfer mainly from the combustion flame to glass and to the furnace wall. To increase the capacity of radiative heat transfer from the flame, we closely studied the atomized heavy oil particle diameter, spraying method and the oxygen introducing method and finally established the basis of our radiative heat transfer type oxy-fuel burner, "Super OFB-LR" [14]. Figure 22 shows the comparison of the flame state between the standard "Super OFB-L" and this new burner, and Fig. 23 shows the comparison of radiative heat flux from the flame. The amount of radiative heat transfer of the radiative burner is more than five times the standard type burner.

The results shown in Table 1 were obtained by applying the radiative heat transfer type oxy-fuel burner to a glass melting furnace. When compared with the conventional air-fuel combustion, this technology is capable of reducing the unit consumption of heavy oil by more than 40% and also reducing NOx by 60%. Furthermore, the quantity of seeds in the glass that could significantly affect the grade of glass has also been successfully reduced by 75%.

Based on the significant results, the first glass melting furnace utilizing oxy-fuel combustion was invented in Japan. After that, the glass industry actively proceeded with the study of the oxy-fuel combustion technology and promoted the introduction of the technology to the production of high value-added type glass.

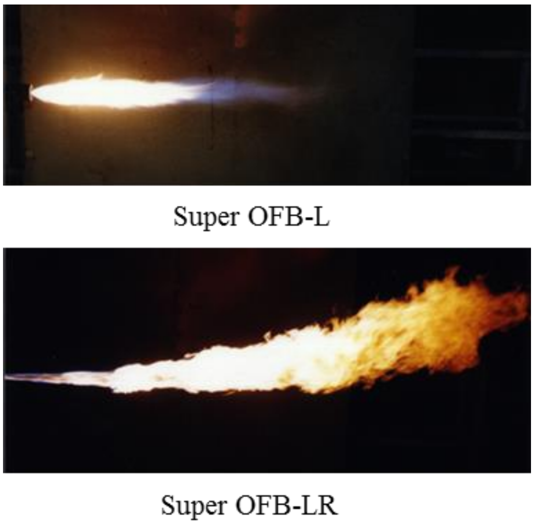


Fig.22 Comparison of oxy-fuel burner flame

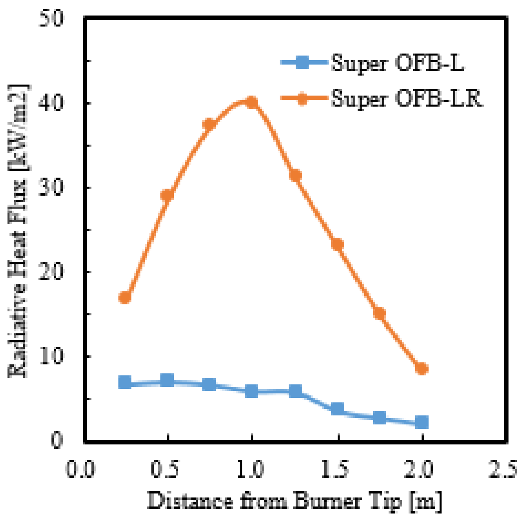


Fig.23 Comparison of radiative heat flux distribution of flames

Table 1 Example of effects of oxy-combustion in a glass melting furnace

	Oxy-combustion	Air combustion
Oil consumption [-]	59	100
Seeds [-]	25	100
NOx emissions [-]	40	100

4.7 Syngas generation technology

The reserves-to-production ratio of natural gas and shale gas is higher than that of crude oil, and those gases can be produced in many countries. Thus, to utilize natural gas and shale gas as energy sources, technologies to produce liquid fuel from those gases (GTL: gas to liquids) have been developed. For that process, it is necessary to convert natural gas etc. to syngas (H_2, CO), and one of such methods is a partial oxidation technique.

We have developed a technology to produce syngas by partially oxidizing gas fuel, such as natural gas, propane, etc., [28, 47]. Figure 24 shows the process flow of the laboratory-scale test equipment.

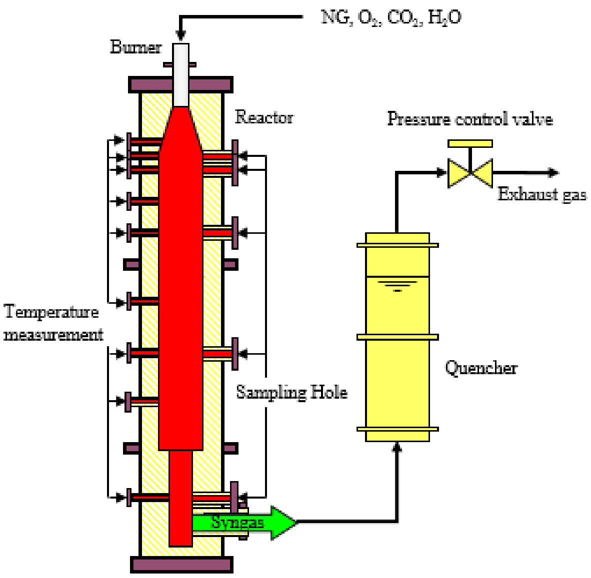


Fig.24 Schematic diagram of test equipment (lab scale)

Using this equipment, we investigated the reaction efficiency, controllability of the H_2/CO ratio of generated syngas, soot generation characteristics, and loads on the combustion chamber, at the furnace pressure between atmospheric pressure and 2.3 MPa. Figure 25 shows an example of temperature distribution in the furnace (gas temperature, temperature of the internal wall of the furnace). As pressure increases, gas temperature near the burner becomes high, and it is possible to increase combustion load by increasing pressure. Table 2 shows an example of attained syngas composition and the equilibrium calculation results. Although unreacted methane remains, the experimental values are consistent with the equilibrium calculation results that take into account the amount of residual methane, and basic data for process design has been obtained. We confirmed that residual methane can be decomposed by installing a catalyst downstream of the reaction furnace.

Based on the laboratory-scale test results, we provided this technology to be used for the syngas generation equipment in the dimethyl ether (DME) direct synthesis project [48] supported by the Ministry of Economy, Trade and Industry. This project uses a demonstration plant that produces 100 tons of DME a day. The syngas generation equipment used in this project generated as much as 14,000 Nm^3/h of syngas (H_2/CO = 1.0) at operating pressure of 2.3 MPa. Through this project, we established a scale-up technology and a long-term stable operation technology.

As this technology is to generate syngas using natural gas, CO_2, and H_2O as raw material, it is possible to contribute to CO_2 recycling by use of recovered CO_2.

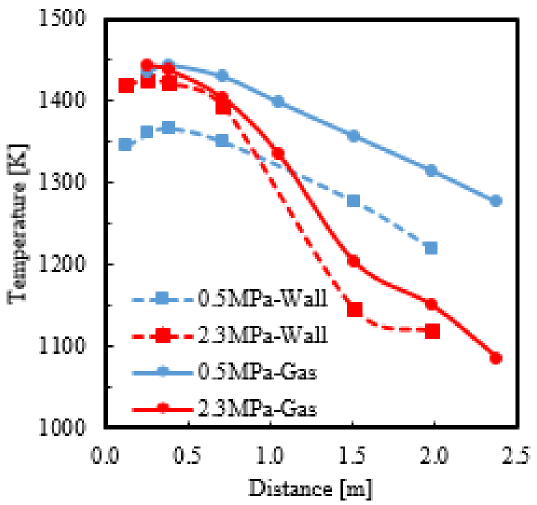


Fig.25 Temperature distribution in the furnace

Table 2 Syngas composition by laboratory test (2.3MPa)

	H_2	CO	CO_2	H_2O	CH_4	Temp. [K]
Measured	22.2	28.1	16.2	22.5	6.2	1312
Equilibrium value	22.6	29.1	17.4	24.3	6.6	1310

4.8 Ammonia combustion technology

Since the oxy-fuel combustion technology can achieve significant energy saving as mentioned above, it can greatly contribute to the reduction of CO_2 emissions. To further reduce CO_2 emissions, large-scale application of renewable energy and expansion of use of carbon-free fuel is necessary. A typical example is hydrogen. However, from the aspect of supplying hydrogen including storage and transportation, liquefaction and high-pressure technologies are required to increase energy density. For this reason, a system has been studied wherein energy is converted to chemical substances (energy carriers) containing hydrogen, transported to consumption locations and stored, and the substances are optimally converted to energy whenever it is necessary. In Japan, ammonia is drawing attention as an energy carrier. Conventionally, a large quantity of ammonia has been used as a raw material of fertilizers and general-purpose chemical goods, and the production method has been established as the Haber-Bosch process. The boiling point of ammonia is 240 K and ammonia liquefies at 0.857 MPa at room temperature, so liquefaction is easier than hydrogen. In addition, a certain scale of ammonia transportation and storage infrastructure already exists at the establishments that own industrial furnaces. Therefore, by using ammonia as a hydrocarbon fuel substitute, it is possible to achieve carbon-free operation.

TNSC was commissioned to develop technology for an ammonia combustion furnace by the Cross-ministerial Strategic Innovation Promotion Program (SIP) under the initiative of the Cabinet Office. Because ammonia contains N in its structure, directly burning ammonia will emit NOx. Based on TNSC's original technologies, by optimizing the nozzle structure and the combustion method, we have developed a technology to enhance flame heat transfer while meeting environmental regulations. Furthermore, with the aim of early application of ammonia mixed combustion to the industrial furnace field, we started to develop degreasing furnace and burner system by utilizing forced convective heat transfer generated by combustion flame in the pretreatment process of the zinc plating steel sheet production (Fig. 26, Fig. 27). We confirmed the availability in the demonstration test performed in the actual zinc plating steel sheet production process [49, 50].

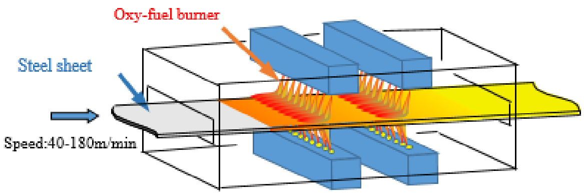


Fig.26 Evaluation test furnace with oxy-fuel burner

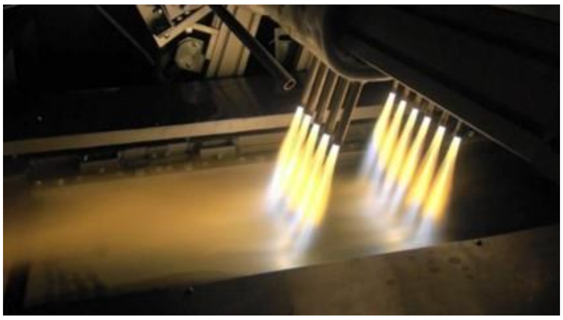


Fig.27 Combustion state of ammonia

5. Prospective view of oxy-fuel combustion

Utilization of oxy-fuel combustion technology began for the purpose of increasing production, and then the use purposes have shifted to energy saving, cost reduction, and to the environmental load reduction. Currently, we believe that the keyword is how to contribute to carbon neutrality by utilizing the oxy-fuel combustion technology.

To do so, it is necessary to develop innovative energy-saving technologies and respond to carbon dioxide recovery and carbon-free energy.

5.1 Innovative energy saving by means of oxy-fuel combustion

As mentioned above, by converting the air-fuel combustion to oxy-fuel combustion, it is possible to reduce the amount of fuel used to almost half. This means that generation of carbon dioxide can be reduced to half when fossil fuel is used. In addition, it is possible to further save energy by using furnaces dedicated for oxy-fuel combustion, recovering waste heat, and using the recovered heat for preheating raw material and oxygen.

Compared with air-fuel combustion, a high-quality combustion field can be attained at very high temperature of approximately 3000 K in the oxy-fuel combustion (as mentioned above). For example, it is effective to use energy in a cascade by combining a process that requires high temperature, a process that is good at medium temperature, and waste heat recovery. To do so, it is necessary to propose and develop new element technologies and processes.

When practically using the oxy-fuel combustion technology, it is necessary to take into account the energy required for producing oxygen and the price of oxygen from the aspect of economy. It is necessary to build a system that can attain energy saving and cost reduction by utilizing waste heat emitted from the processes for the energy to produce oxygen [51, 52].

Thus, combination of oxy-fuel combustion with other technologies will make it possible to save energy in the industrial processes.

5.2 Recovery of carbon dioxide

When performing oxy-fuel combustion of fossil fuel, except for specific gas generated from processes used, the gas emitted after fuel combustion mostly consists of water vapor and carbon dioxide although depending on the fuel components. Water vapor can be easily removed as water during the heat recovery process and highly concentrated carbon dioxide remains. Therefore, it is possible to recover, reuse, and immobilize carbon dioxide (Carbon dioxide Capture, Utilization and Storage: CCUS). If some other gasses are included, use of TNSC's gas separation [53] and purification technologies will facilitate the CCUS process.

In the future, we believe that carbon-free energy used in industrial processes will be mainly composed of carbon-free fuels, such as hydrogen and ammonia, and electricity. However, it is difficult to convert all energy at once in many processes, and conversion will gradually progress by combining various types of energy. As a result, it is necessary to accurately understand the needs and issues concerning the processes and propose suitable oxy-fuel combustion technologies.

6. Conclusion

Basic features of the oxy-fuel combustion, history of TNSC's development of the oxy-fuel combustion technology, and efforts to respond to the market trends have been described above.　In recent efforts to achieve carbon neutrality, the oxy-fuel combustion technology is one of important technologies that can contribute to energy saving.　As mentioned above, by converting air-fuel combustion to oxy-fuel combustion, the unit consumption of fuel can be reduced to almost half. Even though carbon dioxide generated during oxygen production is added, significant energy saving is possible, thereby greatly contributing to the reduction of carbon dioxide emissions.　In the short term, it is possible to significantly save energy only by replacing processes currently operated by air-fuel combustion with oxy-fuel combustion.

Simultaneously, the oxy-fuel combustion can be applied to carbon-free fuels, such as hydrogen and ammonia, and by proceeding with both ways, it is considered possible to contribute to carbon neutrality. We will strive to further improve the technology and develop new technologies by accurately and appropriately responding to market needs.

※スーパーOFB, SUPER OFB, SCOPE-JET, Innova-Jet, CERAMELT are registered trademarks of TAIYO NIPPON SANSO CORPORATION.

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