CO2 Reduction in Asphalt Plants and Development of Ammonia Burner in Nikko Co., Ltd. (2)

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Abstract: Based on the Plan for Global Warming Countermeasures approved by the Cabinet in October 2020, Japan has set a target of a 46 % reduction in greenhouse gas (GHG) emissions from the level of 2013 by the year 2030, and aims to achieve net-zero emission in 2050. Turning our attention to asphalt plants, under these social conditions, emissions from fossil fuels consumed in heating and drying asphalt aggregate in fact occupies 75 % of the CO₂ emitted when producing asphalt mixtures, and of this amount, the energy consumed to evaporate water accounts for more than 50 %. Therefore, moisture content control and pre-drying are effective energy-saving technologies for reducing emissions. Furthermore, by combining a changeover to CO₂-free fuels with these energy-saving technologies, it will be possible to realize a broad reduction in CO₂. Nikko Co., Ltd. is working toward the development of burners that can use various types of these CO₂-free fuels. Among them, ammonia is easier to transport and store than hydrogen, and can also be supplied to asphalt plants located at a distance from industrial zones and urban areas. In addition, because the Haber-Bosch process has already been established as an industrial production process for ammonia, and demand as a CO2-free fuel for thermal power plants is anticipated in the future, it is expected that the distribution volume in Japan will increase and ammonia will become more easily available. To achieve carbon neutrality in asphalt plants, Nikko is engaged in joint research on ammonia combustion with the Akamatsu Laboratory at Osaka University, and has conducted burner combustion tests with outputs up to 100 kW with successful results. In the coming fiscal year, we plan to carry out combustion tests of a burner with an output of 500 kW. We also plan to promote the development of burners with an actual-machine scale output of 5 MW or more in a form that keeps pace with successive advances in the ammonia supply system in Japan, and the development of asphalt plants suitable for that technology. This article reports on the combustion tests carried out from April 2021 to December 2022.

1. Introduction

According to the Intergovernmental Panel on Climate Change (IPCC) Special Report on Global Warming of 1.5 °C released in October 2018, the risk of climate change impacts can be remarkably reduced by limiting the increase in the global average temperature to +1.5 °C from preindustrial levels ¹). Japan ratified the Paris Agreement in 2016, and in October 2020, the Japanese Cabinet approved the goals of reducing greenhouse gas (GHG) emissions by 46 % from the 2013 level by 2030 and achieving net-zero by 2050 ²). In line with the promotion of these environmental initiatives at the global scale, Nikko Co., Ltd. has also begun efforts to reduce CO₂. In 2021, we conducted a survey of Scope 1, 2 and 3 by supply chain emissions calculations, and set targets of reducing CO_2 emissions by 50 % in 2030 and achieving net-zero CO_2 emissions in 2050 ³⁾.

In 2020, CO₂ emissions in Japan totaled 1 044 million t-CO₂/y. Emissions in the industrial sector were 353 million t-CO₂/y, accounting for more than 30 % of total domestic emissions ⁴). In contrast to this, CO₂ emissions from asphalt plants (hereinafter, AP) in Japan in fiscal year 2021 were about 1.3 million t-CO₂/y ⁵), and according to a trial calculation of CO₂ emissions by AP manufactured by Nikko, the emissions of these plants are estimated at approximately 900 000 t-CO₂/y. From the results of the survey conducted in 2021, Scope 1 and Scope 2 emissions in Nikko were 5 056 t-CO₂/y, and the CO₂ emissions from AP manufactured by Nikko

In the breakdown of energy used in AP, consumption of fuels for heating and drying aggregates is 75 %, consumption of electric power for temperature control, machinery drive power, offices, etc. was 17% and heavy machinery fuels and others was 8 % ⁶⁾. The heat sources used for heating and drying of aggregates are fossil fuels, including type A heavy oil and kerosene, which are liquid fuels, and 13A city gas (natural gas with methane as its main component) and LPG (liquefied petroleum gas), which are gaseous fuels. CO₂ emissions from AP can be reduced by 75% by substituting carbon neutral fuels for all of these fuels.

Against this background, up to the present, Nikko has developed burners which can burn high viscosity liquid biomass fuels such as wood tar and waste glycerin and burners capable of burning solid biomass fuels, including carbonized fuels derived from general waste (municipal solid waste)⁷⁾ and crushed rice husks (chaff)⁸⁾.

In the "Green Growth Strategy," which is a government-led initiative to create innovation, the use of hydrogen and ammonia as fuels which do not emit CO₂ when burned has attracted considerable interest, and Nikko has also begun the development of hydrogen burners and ammonia burners that support use of these fuels.

For early social implementation of ammonia burners applicable to AP, Nikko is conducting joint development with the Akamatsu Laboratory of Osaka University, which is considered to be the most advanced organization involved in research on ammonia combustion technology. This article reports on the results achieved in combustion tests of ammonia burners carried out from April 2021 to December 2022.

2. Ammonia Combustion Tests2.1 Properties of Ammonia

Ammonia can be transported and stored more easily than hydrogen, and has attracted attention as a fuel that can be used in large quantities in industrial fields in the future. Because imports from overseas are also transported in the form of liquid ammonia, ammonia also has excellent economic rationality if it can be used in that form.

The thermal properties and combustion characteristics of various fuels are shown in **Table 1**. The combustion velocity of ammonia is slow, being about 1/5 that of

methane, and its flame temperature is also lower than that of methane. Thus, as a fuel, it has the characteristic of being difficult to maintain a flame.

Since ammonia also contains nitrogen, it easily forms fuel NOx when burned. Under Japan' s Air Pollution Control Act, asphalt plants are classified as drying furnaces, and a regulatory value for NOx of 230 ppm by conversion at a 16 % oxygen concentration has been established, except in the Tokyo Metropolitan area and certain other areas. In practice, however, if ammonia burners are to be introduced in AP, acceptance of NOx values exceeding those of existing burners may be difficult.

As an additional issue, because ammonia is toxic, safety countermeasures will be required. These include detoxification of unburned ammonia that remains in the combustion exhaust gas, detection of leaks from the supply system, etc. Based on these considerations, in the development of ammonia burners, new technologies related to "flame stabilization," "low NOx" and "detoxification of unburned ammonia" will be necessary.

Fuel	Ammonia NH3	Propane C3H8	Methane CH4	ydrogen H2
Boiling point at atmospheric pressure (°C)	-33.3	-42.1	-161.6	-252.9
Liquefaction pressure at 20 °C (atm)	8.5	8.5	Normally gas	Normally gas
Lower heating value (MJ/kg)	18.6	46.6	50.2	120.2
Combustibility equivalence ratio range (-)	0.63~1.40	0.51~2.51	0.50~1.69	0.10~7.17
Maximum combustion velocity (m/s)	0.07	0.43	0.37	2.91
Ignition point ($^{\circ}\!$	651	432	537	500
Maximum adiabatic flame temperature (°C)	1750	2020	1970	2120

Table 1 Thermal properties and combustioncharacteristics of various fuels 9)

2.2 Overview of Ammonia Burner

The main stream in AP operating in the suburbs of urban areas is machine types with an aggregate drying capacity of 30 t/h in small units to 120 t/h in large-scale facilities. In actuality, the outputs of burners suitable for these AP range from 3 MW to 12 MW, and burners with different outputs are necessary for each machine type.

As the fuels used with these burners, with the exception of urban areas where city gas is supplied, the large

majority are operated with type A heavy oil. Figure 1 shows the structure of an oil burner manufactured by Nikko. As shown in the figure, the fuel oil is pressurized by a booster pump and injected through a high pressure atomizing nozzle. The atomized fuel oil is caught in the negative pressure region formed on the front side of the flame stabilizer by the flow of combustion air, and forms "fireballs", i.e., the flame in the recirculation zone. With these "fireballs" as a heat source, the fuel oil injected from the nozzle is successively gasified and burned. At the same time, the flow velocity of the combustion air discharged from the periphery of the flame stabilizer is increased by increasing the static pressure of the combustion air supplied by the fan, which forms a turbulent flow and promotes mixing and agitation of the gasified fuel oil and combustion air in the mixing chamber. This is done to enhance combustion efficiency and shorten the flame length by increasing the combustion velocity. Therefore, as shown in Figure 2, a combustion chamber is not necessary in the AP because the burner is attached directly to the end of the drying kiln, and heating and drying of the aggregate are performed by radiant heat transfer from the flame formed inside the kiln and contact heat transfer from the hot blast.



Figure 1 Structure of oil burner manufactured by Nikko Co., Ltd. ¹⁰⁾



Figure 2 Burner and drying kiln of AP ¹¹)

The characteristics of conventional burners described above should also be retained in ammonia burners for use in AP, which is the target of this development. However, in order to identify uncertainties in the initial stage of development and collect various types of data, a combustion chamber was purposely provided at the leading end of the burner, as shown in Figure 3, and a method of mixed firing with type A heavy oil, in which ammonia is supplied in the combustion chamber, was adopted. Moreover, as also mentioned above, to adapt an ammonia burner to AP, ammonia combustion technologies that can satisfy regulatory values without additional denitrification equipment or equipment to detoxify unburned ammonia in the AP are necessary.

As countermeasures for unburned ammonia, we studied increasing the combustion efficiency of ammonia, which has a slow combustion velocity, with a fuel that has a higher combustion velocity than ammonia and can form a stable flame. At present, most fuels used with AP, with the exception of some city gas, are liquid fuels such as type A heavy oil. Therefore, considering easy introduction in fuel substitution, a mixed fuel firing test with type A heavy oil was prioritized.

Although direct-type deodorization equipment which decomposes the odorous components formed from recycled materials is installed in AP, the retention time is insufficient for treating ammonia due to its slow combustion velocity. Thus, it would be difficult to use this equipment as a detoxification device for unburned ammonia.

To reduce NOx, we adopted the two-stage combustion method, which is already generally established as a NOx countermeasure technology. Two-stage combustion is a method in which CO is formed in the first stage by reducing the excess air ratio to 1 or less, and this CO is used to reduce NOx and return it to N2. Then, in the second stage of combustion, the unburned portion is burned by supplying the deficient air. In addition to the unburned ammonia detoxification effect, an effect of suppressing thermal NOx by reducing local high-temperature hot spots can also be expected because slow combustion with deficient air prevails. Figure 3 shows the position where air is supplied for second-stage combustion.



Figure 3 Structure of test burner

2.3 10 kW and 100 kW Combustion Test Equipment

When using the ammonia combustion test equipment of the Akamatsu Laboratory at Osaka University, the available output range for tests is 10 kW and 100 kW. However, the tests were deliberately conducted beginning with the small 10 kW scale unit in order to identify other uncertainties such as lack of experience in handling of ammonia, which is a harmful chemical, treatment of unburned ammonia, suppression of fuel NOx precipitation, improvement of inadequate flame stability caused by the slow combustion velocity of ammonia, etc. Based on the results with this small-scale equipment, tests were carried out at the 100 kW scale, i.e., scaled up by a factor of 10. The flowchart of the combustion test equipment used in this development is shown in Figure 4. In both the 10 kW and 100 kW combustion test equipment, a furnace with an inner ceramic construction is installed, and measurement of the temperature distribution in the furnace and exhaust gas analysis are possible. In addition, the exhaust gas from the furnace is discharged into the atmosphere by way of a detoxification furnace in order to remove unburned ammonia, if generated, and a cooling tower which cools the exhaust gas. Sight holes of heat-resistant glass specification are installed at various points on the combustion test equipment so that the flame can be confirmed. Figure 5 shows the condition of a combustion test with the output of 100 kW.

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— 4 —



Figure 4 Flowchart of 10 kW and 100 kW combustion equipment



Figure 5 Condition of 100 kW combustion test

2.4 Combustion Exhaust Gas Measurement Devices

Measurements of the ammonia combustion exhaust gas were carried out during this combustion test. The measurement items and measurement methods are shown in **Table 2**. The measurements were conducted continuously at the outlet of the test burner and various points on the furnace. Because ammonia has an action that reduces NOx, a measurement device (Horiba, Ltd., MEXA1400QL-NX) which is capable of simultaneously measuring both NOx and ammonia by using a quantum cascade laser (QCL) was employed to investigate the interrelationship of NOx and ammonia ¹²⁾. Among the various NOx compounds, tests were conducted focusing on N2O (nitrous oxide), as this compound has a greenhouse effect (warming potential) approximately 300 times more powerful than CO2. All values of NOx in the following are converted values for an oxygen concentration of 16 %.

Table 2 List of measurement items and measurement methods

Measurement item	Measurement method
02	Zirconia oxygen concentration cell type O ₂ analyzer
	Dumbbell type paramagnetic O2 analyzer
CO, CO ₂	Nondispersive infrared absorbance (NDIR) method
NO _x	Vacuum type chemiluminescence detector (CLD)
NO, NO ₂ , N ₂ O, NH ₃	Quantum cascade laser-infrared spectroscopy

2.5 Combustion Test Parameters

The parameters (factors and levels) implemented in this combustion test are shown in Table 3. From the table, the burner output (kW) was set to the four levels of 10 kW, 75 kW, 100 kW and 125 kW. Burners with the two outputs of 10 kW and 100 kW were used, and the combustion tests for the three levels of 75 kW, 100 kW and 125 kW were carried out with the burner with the output of 100 kW. The mixed fuel firing ratio (%) was defined as the ratio of the heating value of ammonia to the total heating value including both the type A heavy oil and ammonia. The combustion tests were conducted in the range of levels from 10 % to 70 %. For the excess air ratio (-), the two levels of excess air ratio A and excess air ratio B using the furnace outlet as a standard are selected. The relationship 1.0 < excess air ratio A < excess air ratio B is materialized by these excess air ratios. The secondary air ratio (-) indicates the ratio of air supplied in the second stage of two-stage combustion, and the three levels of secondary air ratios 0, a and b are selected. These secondary air ratios materialize the relationship 0 < airratio a < air ratio b. Needless to say, the secondary air ratio of 0 means one-stage combustion. Although not shown in the table, a primary air ratio equivalent to the first stage of two-stage combustion also exists, and the relationship excess air ratio = primary air ratio + secondary air ratio is materialized between the primary and secondary air ratios.

Table 3 List of combustion test parameters

Burner output	Mixed fuel	Excess	Secondary
(kW)	firing ratio (%)	air ratio (-)	air ratio (-)
10, 75, 100, 125	10 to 70	A∼B	0, a, b

3. Combustion Test Results and Discussion3.1 Results of 10 kW Combustion Test andDiscussion

Figures 6 and 7 show the condition of the flame with ammonia mixed fuel firing ratios of 10 % and 30 % in the burner with the output of 10 kW, under conditions in which the output and excess air ratio were fixed. As can be confirmed from these figures, in comparison with the flame with the 10 % ammonia mixed firing ratio, with the ratio of 30 %, a pale-colored flame has spread through the entire combustion chamber, and the flame length is also longer due to the increase in hard-to-burn ammonia. Here, the whitish-appearing region of high luminescence at the right edge of the figure seems to be the flame of the type A heavy oil. Based on these facts, it is thought that the type A heavy oil, which is a liquid fuel, burns first, and the gaseous ammonia then burns following that, showing the very slow combustion velocity of ammonia. It is known that the combustion velocity of gaseous fuels is generally faster than that of liquid fuels.



Figure 6 Flame with 10 % ammonia mixed fuel



Figure 7 Flame with 30 % ammonia mixed fuel

Next, **Figure 8** shows the values of NOx in the exhaust gas due to the difference in one-stage combustion and two-stage combustion for ammonia mixed fuel firing ratios of 30 % and 50 %. The figure shows that NOx is higher than 300 ppm with one-stage combustion, but decreases to 200 ppm or less with two-stage combustion. In both one-stage combustion and two-stage combustion, a tendency in which the higher ammonia mixed firing ratio shows lower NOx values can be seen, and even when the ammonia mixed firing rate reaches 70 %, a stable flame effect was obtained and unburned ammonia could not be detected. From these results, the tendency and basic data concerning the NOx concentration could be obtained for parameters such as the excess air ratio, mixed fuel firing ratio, effect of two-stage combustion, etc. in combustion tests with an output of 10 kW.

Figure 8 NOx in exhaust gas in one-stage combustion and two-stage combustion with different mixed fuel firing ratios (10 kW)



3.2 Results of 100 kW Combustion Test and Discussion

Combustion tests were conducted at the output levels of 75 kW, 100 kW and 125 kW to investigate the differences between one-stage combustion and two-stage combustion. Figure 9 shows the NOx values when the excess air ratio was changed from A to B in these tests. However, the mixed fuel firing ratio is 70 %, and the secondary air ratio in two-stage combustion is fixed. As in the combustion tests with the 10 kW output, a tendency in which NOx decreases when two-stage combustion is performed can also be confirmed from this figure. Although limited, conditions which can reduce NOx to the same level as in city gas firing were discovered. Moreover, the generally-known tendency that the NOx concentration rises as a result of an increased excess air ratio and output was also confirmed with both one-stage combustion and two-stage combustion.



Figure 9 Tendency of excess air ratio and NOx with various output levels

On the other hand, formation of N₂O (nitrous oxide) by ammonia combustion is a concern. Here, a level of about 10 to 15 ppm was observed with 75 kW, but at 100 kW and 125 kW, N₂O did not exceed 1 ppm. It is known that the formation of N₂O in fossil fuel combustion is strongly dependent on the combustion temperature, and the value of N₂O decreases in high-temperature combustion ¹³. Because the amount of N₂O formation tended to increase at lower outputs, i.e., lower temperatures, was also confirmed in this combustion test, it will be necessary to pay attention to changes in the amount of N₂O formation in future combustion tests.

Next, **Figure 10** shows the values of NOx when the secondary air ratio is changed in two-stage combustion. At the higher secondary air ratio (secondary air ratio a < secondary air ratio b), the reduction of NOx in two-stage combustion increased, and a decrease in NOx could be observed.



Figure 10 Relationship of secondary air ratio and NOx

3.3 Verification by Simulation Software

Combustion of ammonia was simulated by a simple model of the 100 kW test burner using the simulation software STAR-CCM+ (manufactured by Siemens). Figure 11 shows the distribution of unburned ammonia when a simulation was performed under a certain two conditions. For comparison with actual combustion, combustion was performed under conditions approximating those of the simulation, and when the ammonia was actually measured at points 1 to 4 in each figure, an ammonia level of about 500 ppm was detected only at point 3, confirming that the simulation results were consistent with the actual measurements. It is thought that further improvement in the accuracy of simulations by this STAR-CCM+ program will contribute to optimization of the design of combustion chambers, etc.



Figure 11 Simulation of unburned ammonia

4. Issues for Introduction in AP and Future Prospects

Various issues remain for installation of the ammonia burner in actual AP. Of course, it will be necessary to confirm reproducibility when the test burner used in these combustion tests is upscaled to actual machine size, and develop combustion technologies for achieving lower NOx. In addition to these points, in many cases, continuous operation of AP burners for 3 to 4 hours is performed intermittently. As a result, numerous parts are subjected to changes such as rapid heating and rapid cooling, and an output range with a turndown ratio of about 1 : 5 is required, corresponding to changes in the feed rate and condition of the materials to be dried. Since the output was varied from 75 kW to 125 kW with the 100 kW test burner in these combustion tests, it can be said that turndown could be confirmed to 1 : 1.6. Here, the turndown ratio of a burner indicates the possible range of changes in burner output, and is expressed by the ratio of the minimum and maximum outputs.

In the next fiscal year, we plan to conduct combustion tests using a burner with a 500 kW output, which is closer to the actual machine scale, and we hope to solve the above-mentioned issues in these combustion tests. Because a burner with an output of 500 kW can be used as a burner for the drying kiln of the Nikko product NMD-100¹⁴⁾ as an application other than AP, we also intend to promote the development of ammonia burners to use in non-AP applications. If necessary, we will also study the development of denitrification equipment and detoxification furnaces for those applications.

Moreover, depending on future trends in carbon neutral fuels, it is also possible that fuel conversion for the type A heavy oil in AP that currently use type A heavy oil may proceed in the direction of city gas, which can reduce CO_2 by 25 %, or propane, which can achieve a 15 %

reduction. For this reason, we are currently conducting mixed fuel firing tests with gases in parallel with mixed fuel firing tests with liquid fuels.

5. Conclusion

Combustion tests up to a burner output of 100 kW were conducted with the ammonia combustion test equipment of the Akamatsu Laboratory of Osaka University. These combustion tests confirmed that mixed fuel firing with a fuel which has a faster combustion velocity than ammonia, and thus can form a stable flame, and two-stage combustion are effective as new technologies for "flame stabilization," "low NOx" and "detoxification of unburned ammonia," which are required in ammonia burner development. Although limited, conditions which can lower NOx to the same level as in city gas combustion were also discovered. In addition, the results of a simulation using STAR-CCM+ were compared with actual measurements, and qualitative consistency was obtained. Therefore, use of simulations is expected to contribute to design optimization in the future.

At present, the authors are also conducting mixed fuel firing tests with gaseous fuels in parallel with mixed fuel firing tests with liquid fuel, and in the coming fiscal year, we aim to develop a burner that can be introduced in actual AP using a burner with 500 kW output, which is closer to actual machines.

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