Development of Energy-Saving Oil-Contaminated Soil Remediation Plant, Its Transition and Future Prospects

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1. Introduction

Through the post-war era of high economic growth, Japan grew to become a major economic power with a GDP surpassing even that of the United States during the "bubble economy" period of the late 1980s. As a result of this economic growth, the Japanese people enjoy affluence and convenience. This material abundance and convenience were achieved by mass production, mass consumption and mass disposal of industrial products, and as the price of this, future generations have been left to bear the problems of pollution and environmental destruction, as well as global warming.

In response to the problem of soil pollution, Japan enacted the Soil Contamination Countermeasures Law, effective February 15, 2003. This was more than 10 years later than similar laws in Europe and the United States, and was also the most delayed among laws for the seven typical types of pollution. The purpose of the laws concerning the other six types of pollution (air pollution, water pollution, noise, vibration, offensive odor and ground subsidence) which had already been enforced was to prevent the occurrence of pollution and spread of damage in advance by regulating the concentration and amount of discharges of byproducts and waste accompanying economic activity. In contrast to those laws, the most important purposes of the Soil Contamination Countermeasures Law were firstly to understand the actual condition of soil contamination and act to prevent adverse impacts on human health due to consumption of groundwater, direct ingestion of contaminated soil, etc. For this reason, the law was named the "Soil Contamination Countermeasures Law," and not the "Soil Contamination Prevention Law."

The Soil Contamination Countermeasures Law specifies 27 types of designated hazardous substances as environmental standards for soil established by the Japanese government. The total of 27 designated hazardous substances specified by the law consists of 12 types of volatile organic compounds (VOC) as Class 1, 10 heavy metals as Class 2 and 5 types of agrichemicals and PCB as Class 3 designated hazardous substances.

The Class 1 designated hazardous substances include chlorinated organic compounds (COC) such as trichloroethylene ($CHCl=CCl_2$, soil leachate standard: 0.03 mg/L), tetrachloroethylene ($CCl_2=CCl_2$, leachate standard: 0.01 mg/L), dichloromethane (CH_2Cl_2, leachate standard: 0.02 mg/L), etc. Because these COC are organic compounds whose molecules contain chlorine atoms, are chemically stable and are nonpolar molecules, etc., they are used as solvents in degreasing, cleaning and dry cleaning of electronic components, and other applications. As their effects on the human body, these compounds cause paralysis of the central nervous system, and if inhaled, symptoms may include dizziness, headache, sleepiness, nausea, difficulty in speaking or walking, unconsciousness, etc. Among physical properties, they are virtually insoluble in water, and their density is greater than that of water. Since their boiling point is 100℃ or less under normal pressure, they can be desorbed easily from soil by heating to increase the vapor pressure. The hot soil method, in which contaminated soil is heated by the heat of hydration of quicklime added to the soil, is known as one soil remediation method. This is a desorption method whereby VOC are removed from soil by using the physical change (liquid → gas) caused by the change in the temperature of the VOC. Other remediation methods such as chemical decomposition and microbial decomposition have also been applied practically to Class 1 substances.

The Class 2 designated hazardous substances include heavy metals such as mercury (Hg; soil concentration standard: 15 mg/kg, leachate standard: 0.005 mg/L), cadmium (Cd; concentration standard: 150 mg/kg, leachate standard: 0.01 mg/L), lead (Pb; concentration standard: 150 mg/kg, leachate standard: 0.01 mg/L), hexavalent chromium (Cr^{+6}; concentration standard: 250 mg/kg, leachate standard: 0.05 mg/L), etc. Among their effects on the human body, the Japanese government recognizes Minamata disease caused by mercury and itai-itai disease caused by cadmium as pollution-related diseases. Minamata disease is a toxic central nervous system disease caused by methyl mercury, with symptoms that include ataxia (loss of muscular coordination), sensory disorder, hearing impairment, etc. The main symptoms of itai-itai disease are multiple renal tube dysfunction (kidney damage) and osteomalacia (softening of the bones).

These heavy metals are considered to exist in soil in the state of compounds such as oxides, hydroxides and sulfides. In the presence of groundwater, it is known that they are eluted due to the effect of the hydrogen ion concentration of the groundwater and also exist in the ionic state of the simple atoms. Because these heavy metals are harmful even in the atomic or independent ionic states, they cannot be detoxified by the decomposition methods used with Class 1 substances.

Because many heavy metals have high melting points and boiling points, if a remediation process based on physical properties (change of the three states of matter, etc.) is employed as a method for removing heavy metals from soil, an energy-intensive purification process will be necessary, resulting in increased remediation costs. Therefore, focusing on the fact that many compounds of heavy metals adhere to the surface of soil particles, one effective method for reducing the concentration of heavy metals in soil is the washing method, in which silt and sandy soils, which have large specific surface areas, are removed from the soil by classification.

The Class 3 designated hazardous substances include PCB (polychlorinated biphenyl; leachate standard: less than the detection limit) and pesticides and herbicides such as thiuram (tetramethylthiuram disulfide; leachate standard: 0.006 mg/L) and simazine (6-chloro-N,N-diethyl-1,3,5-triazine-2,4-diamine; leachate standard: 0.003 mg/L). Because the PCBs have been designated as strongly toxic carcinogens and are persistent compounds, the Japanese government enacted the Act on Special Measures Concerning Promotion of Proper Treatment of PCB Wastes in connection with the handling of PCBs, planning to complete detoxification treatment by March 31, 2023. Since these Class 3 substances are organic compounds, detoxification is possible by thermal decomposition under appropriate temperature conditions.

On the other hand, it is no exaggeration to say that petroleum became a driving force for high economic growth in Japan following the Second World War. Petroleum is not only as primary energy source, but is also as a raw material for chemical products and has excellent economy, safety and handleability, and therefore was used in large quantities as a cleaning solution and solvent in the manufacturing processes of industrial products, and was also disposed of as waste in large quantities. As the price for this, we have now reached the final stage where future generations will bear the negative legacy of oil-contaminated soil, as shown in **Photo-1.**
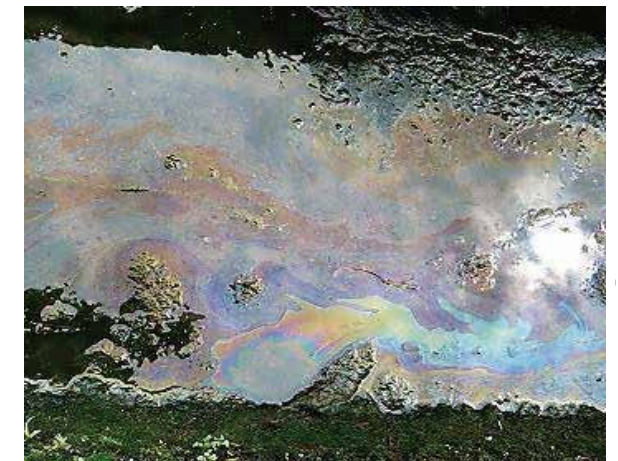

Photo-1 Oil-contaminated soil 1)

Oil-contaminated soil is frequently detected from former factory sites where production activities were carried out, and it is said that more than half of all soil contamination occurs by way of oil. Soil contamination by oil also causes serious problems in the living environment in the forms of oily odor and oil film, as can be seen in **Photo-1**. Nevertheless, oil-contaminated soil, which is the target of remediation in this report, has not been classified as a designated hazardous substance under the Soil Contamination Countermeasures Law. Where oil contamination of soil is concerned, according to the environmental standards for soil established by the Japanese government, benzene (C_6H_6; leachate standard: 0.01 mg/L, which is a constituent component of oil, is

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only an object of regulation as a Class 1 designated hazardous substance. Moreover, because benzene displays carcinogenicity, it is no longer used as an organic solvent.

In this connection, in March 2006, the Soil Contamination Technical Standards Expert Committee of the Soil Agricultural Chemical Committee of Japan's Central Environmental Council released a report entitled the "Guidelines for the Countermeasures against Oil Contamination – Landowner's Response Policy Concerning Oil Slick and Odor Problems from Mineral Oil Contaminated Soil." [1] (hereinafter, the "Guidelines"), which summarize the point of view on the response to oil contamination problems and the matters to be used as reference when carrying out surveys and implementing countermeasures at sites where oil contamination problems have occurred. Although the biggest goal of the Soil Contamination Countermeasures Law is to prevent adverse impacts on human health, the Guidelines differ greatly in that their purpose is to remove obstacles to preservation of the living environment.

From the viewpoint of the history of remediation measures for oil-contaminated soil, remediation measures were taken voluntarily on a private-sector basis before enforcement of the Soil Contamination Countermeasures Law. This was done as a risk-avoidance action to reduce the potential decrease in the appraised value of land due to contamination when selling land. Bioremediation using the healing power of nature and remediation employing cement kilns, in which high-temperature incineration technology is applied, are known as remediation technologies for oil-contaminated soil, but these technologies have not been widely adopted in Japan due to concerns about reliability, economy, etc. Based on these needs of the times, the establishment of a soil remediation technology that enables economical, reliable, speedy remediation of oil-contaminated soil, detoxification of the oil by thermal decomposition, and early restoration of microorganisms and plants inhabiting the soil after remediation, while also maintaining its nature as soil without impairing its essential functions, has long been desired.

This report traces the evolution of oil-contaminated soil remediation systems developed by the authors, presents an overview of a newly-developed energy-saving oil-contaminated soil remediation system, and introduces its energy-saving features, purification performance and

future technological prospects.

Photo-2 Capping oil-contaminated soil with asphalt pavement [12]

Photo-3 Cut-off wall (water barrier wall) [1]

2. Past Remediation Technologies for Oil-Contaminated Soil

Countermeasure technologies for oil-contaminated soil in the past can be broadly divided into spread prevention measures that attempt to prevent damage from reaching humans and animals by preventing the spread of oil, and remediation measures that actively remove oil from soil and recover or detoxify it by decomposition.

In spread prevention measures for oil contamination, when no permeation and diffusion of the oil into the groundwater occur, it is necessary to carry out oil film capping/oil odor capping at the ground surface. In this case, the most economical methods are banking and paving. Banking involves covering the oil-contaminated soil with soil from above, and can shield against oil odor and oil films. Paving includes asphalt pavement, as shown

in **Photo-2**, concrete pavement and others. In comparison with banking, paving has a superior shielding effect.

When oil permeation and diffusion into the groundwater occur, it is necessary to take measures to prevent the oil from spreading in the groundwater. In this case, a cut-off wall (impermeable water barrier), as shown in **Photo-3**, barrier wells, etc. are effective. A cut-off wall is a measure in which the spread of oil is prevented by driving sheet piles to a water-impermeable layer so as to surround the area where oil may leak out. In the barrier well method, the spread of oil is prevented by digging pumping wells around the perimeter of oil leaked area. The oil contained in the pumped-up water is removed, and the purified water is then discharged naturally. As a risk assessment system for permeation and diffusion of oil into groundwater, Sakamoto et al. conducted an analysis of the phenomenon of oil movement in groundwater and a quantitative assessment of the health risk to humans [2)-5)].

On the other hand, remediation technologies for oil-contaminated soil can be broadly classified as soil excavation and removal methods (ex-situ methods) and in-situ remediation methods. Purification methods for excavated oil-contaminated soil include thermal treatment, soil washing, bioremediation, use as a raw material for cement and others. Hashimoto et al. [6] presented a detailed report on actual examples of on-site remediation technologies for soil contaminated with gasoline, diesel fuel, kerosene, etc., which is most common at the former sites of gas stations in urban areas, etc. In these methods, the contaminated soil is removed by excavation, and is then backfilled to its former position after purification treatment at the contamination site.

Thermal remediation is a method by which oil is desorbed from soil by heating the soil in order to increase its vapor pressure. However, almost all soil contains moisture, which consumes thermal energy in the heating process, and this is an obstacle to the heating process. Thermal desorption methods can be classified according to the soil heating temperature as drying-heating treatment methods, in which the oil is volatilized and separated from the soil at 200 to 400 ℃, thermal desorption treatment methods, which desorb the oil from the soil at 400 to 600 ℃, and thermal decomposition treatment methods, which decompose the oil content at 600 to 1 000 ℃. Furthermore, thermal remediation

methods for contaminated soil are also classified as continuous heating methods and batch heating methods, and the continuous methods are further classified as the counterflow type or the parallel flow type, depending on the relative flow directions of the soil being treated and the hot air used as the heat medium. In the counterflow type, the contaminated soil and hot air flow in opposite directions, while in the parallel type, they flow in parallel. The series of oil-contaminated soil purification systems described in this paper fall under the two treatment methods of heating-drying, in which the soil heating temperature is in the range of 200 to 400 ℃, and thermal desorption at 400 to 600 ℃. Both are continuous heating methods.

Soil washing is a method in which the concentration of the oil contained in the soil is reduced by classifying the soil by using a solvent, surfactant or the like. It is effective when used in combination with bioremediation or other appropriate methods. Due to the basic principle of reducing the oil concentration by classification of the soil, it is difficult to adapt to soils with high silty or clayey contents.

Bioremediation after excavation and removal is a purification method which utilizes the oil decomposition capacity of bacteria or other microorganisms. Methods which have reached practical application include biostimulation, in which decomposition of the oil is accelerated by supplying nutrient salts to oleophilic bacteria (oil-eating microbes: OEMs) living in the oil-contaminated soil, and bioaugmentation by injecting microorganisms with a high oil decomposition capacity into the oil-contaminated soil.

Bioremediation has the advantage of a lower remediation cost in comparison with other remediation methods. It is also the method the friendliest to the natural environment, as it utilizes the natural healing power of the soil. However, as one limitation, it is difficult to adapt to the winter season, when the activity of microorganisms decreases. Mori et al. [7] conducted research on the injection amount and injection rate of nutrient salts in bioremediation, and reported that maintaining the nutrient salts in an unsaturated state in the soil is more effective. Suto et al. [8] investigated the effect of applying quicklime treatment to oil-contaminated soil on oil-decomposing bacteria living in the soil. According to this report, it was found that the density of bacteria in the

soil decreased, but bacteria survived even though the pH of soil the shifted to around 12 or more due to addition of quicklime, and the surviving bacteria contributed to decomposition of oil that remained after the quicklime treatment. When bioaugmentation is to be applied, safety must be studied in advance in accordance with "Guidelines on Use of Microbial-based Bioremediation" (March 30, 2005; joint notification by the Ministry of Economy, Trade and Industry and the Ministry of the Environment). In a safety-related study, Kitayama et al. [9] conducted chronic toxicity and acute toxicity tests with mice for bacteria that are effective in decomposition of oil and confirmed the safety of these bacteria for outdoor use.

Use of contaminated soil as a raw material for cement is a method in which oil-contaminated soil which has been excavated and removed is transported to a cement production facility and effectively utilized as a raw material for cement. Effective utilization of all of the components as raw material for cement is possible, including calcium (Ca), silica (SiO_2), alumina (Al_2O_3) and iron (Fe), as these are chemical components of cement, and soil also contains large amounts of the same substances. However, there is a limit on the amount of oil-contaminated soil that can be accepted, as the blending ratio of these chemical components is an important element for maintaining cement quality.

Next, in-situ remediation techniques include methods such soil vapor extraction (SVE), in-situ bioremediation and chemical oxidation treatment.

Soil vapor extraction is a vacuum extraction method in which volatile components (VOC) are volatilized by reducing the pressure of oil that exists underground, and then extracting and removing those compounds. Its range of application can be expanded by combined use with other techniques such pumping-up groundwater, air sparging (underground aeration), etc.

In-situ bioremediation can be adapted to cases where excavation is impossible due to the presence of buildings or the cost of excavation would be prohibitive due to the depth of the contamination. In this remediation method, decomposition of the oil content is accelerated by injecting a nutrient salt (biostimulation) or a microbiological agent (bioaugmentation).

Chemical oxidative decomposition is a remediation method based on oxidative decomposition of organic compounds, including oil, when exposed to a large

quantity of oxygen, which is generated by reacting an oxidant such as hydrogen peroxide using iron as a catalyst. Oxidative decomposition reduces the oil to harmless CO_2 and water.

Before application of any of these remediation methods, it is necessary to carry out a treatability test using actual contaminated soil and fully study the effectiveness of the method and the environmental impacts of the components that will be formed as byproducts.

3. Transition of Nikko Thermal Remediation Systems for Oil-Contaminated Soil

3.1 Overview

Nikko Co., Ltd. (hereinafter Nikko) developed and introduced the Nikko Thermal Remediation system (hereinafter, NTK system) into the market in 1999. The NTR system is based on technologies which Nikko developed for its asphalt plant aggregate drying systems and direct combustion-type deodorization system. In response to the needs of society, the company has also successively improved the purification functions adapted to increasingly diverse pollutants, and now offers a NTR Series product lineup consisting of six types, from NTR-Stage I to NTR-Stage VI, which differ in terms of the contaminants targeted for remediation, treatment functions, and whether the system includes wase heat recovery or not [10)-15)]. **Table-1** shows the lineup of the NTR Series and the features, thermal treatment temperatures, and remediation targets of each system.

The systems in the NTR Series perform thermal remediation treatment of oil-contaminated soil in a temperature range from 150 to 600 ℃, enabling sure remediation of large volumes of contaminated soil at low cost. In addition, because thermal remediation is performed below the melting point of the soil, the essential properties of the soil are unchanged, and because harmful substances are decomposed and detoxified within the system, secondary pollution is not a concern. These are important features of the NTR Series.

Internal heating type rotary kilns are used as the primary treatment device. In the NTR Series, the counterflow type is used in NTR-Stage I and Stage II, and the parallel flow type is adopted in Stage III to Stage VI.

Table-1 Lineup of Nikko Thermal Remediation systems (NTR systems) and substances targeted for purification

Plant type		Stage I	Stage II	Stage III	Stage IV	Stage V	Stage VI
Soil heating treatment temperature (℃)		150～250	150～300	150～400	150～600	150～1,000	150～1,000
Primary treatment device exhaust gas temperature (℃)		120～180	120～180	200～500	200～800	200～1,200	200～1,200
Secondary treatment temperature (℃)		～800	～800	～800	～1,200	～1,200	～1,200
Tertiary treatment temperature (℃)		－	200～350	－	－	－	－
Tertiary treatment exhaust gas temperature (℃)		－	250～400	－	－	－	－
Cooling tower		－	－	○	○	○	○
Heat exchanger		○	○	－	－	○	○
Preheating kiln		－	－	－	－	－	○
Types of oil contaminants	Gasoline (equivalent)	○	○	○	○	○	○
	Kerosene (equivalent)	○	○	○	○	○	○
	Diesel (equivalent)	△	○	○	○	○	○
	A type heavy oil (equivalent)	－	○	○	○	○	○
	Machine oil	－	○	○	○	○	○
	C type heavy oil (equivalent)	－	△	○	○	○	○
	Tar/pitch	－	－	○	○	○	○
Class 1 Designated hazardous substances, VOC		－	－	○	○	○	○
Mercury, cyanogen compounds, agrichemicals		－	－	－	○	○	○

Note)○ : Applicable, △: Applicable with conditions, － : Not applicable.

The former counterflow type is a method in which the hot air that functions as the heat medium and the soil being heated flow in opposite directions in the rotary kiln. It is an effective method for soil treatment when the remediation target is light crude oil which has a comparatively low boiling point and large-volume treatment is to be performed.

In the latter parallel type, the hot air and soil flow in the same direction. This is an effective method when treating remediation targets that have high boiling points and are hard to decompose.

Corresponding to the thermal treatment temperature of the soil, a rotary kiln with a metallic structure without refractory lining is used in the NTR systems from Stage I to Stage III, while a rotary kiln with a structure that includes an inner lining of a refractory castable with excellent heat resistance and heat storage properties is installed in the NTR systems from Stage IV to Stage VI. The Stage VI NTR system is equipped with an external heating-type preheating kiln utilizing waste heat for pre-drying of the soil in the process before the primary treatment device to achieve thoroughgoing energy saving.

All NTR systems are equipped with a secondary treatment device for detoxification of the oil components desorbed from the soil by oxidative decomposition, and a

humidity control drum with the combined functions of cooling the thermally-treated soil, dust prevention, and returning the soil to its original moist condition.

Although the secondary treatment devices differ depending on the type of contaminant, all of these devices secure the holding time necessary to complete the oxidative decomposition reaction of the organic compounds in the temperature range of 750 to 1 200 ℃, which is required for this reaction. With the exception of Stage III of the system, all secondary treatment devices are equipped with heat exchangers to achieve energy saving by recovering waste heat.

In the latter device, i.e., the humidity control drum, a moist condition is achieved by directly sprinkling water on the thermally-treated soil, but because a large quantity of steam and accompanying dust forms in this process, the drum is also equipped with a dust collector to capture this dust.

In addition to the above, the Stage III systems and higher, which are suitable for hard-to-decompose contaminants that require a high treatment temperature, are equipped with stock bins to accelerate remediation by oxidizing and aerating the oil that remains inside the soil particle by equalizing the heating temperature of the soil.

The following presents an overview of the six types from

NTR-Stage I to NTR-Stage VI, and mentions their respective features.

3.2 NTR-Stage I

NTR-Stage I was the first remediation system for oil-contaminated soil developed by Nikko. As shown above in **Table-1**, the target of remediation is soil contaminated with light crude oils such as gasoline and kerosene. The targeted light crude oils are equivalent to oils whose main components are aliphatic hydrocarbons and aromatic hydrocarbons with carbon numbers of C_5 to C_{18}, and have boiling points in the range from room temperature to 250 ℃. **Figure-1** shows the process flow of NTR-Stage I, and **Photo-4** shows a general view of a plant. The following presents an overview of the soil remediation process based on the above-mentioned figure.
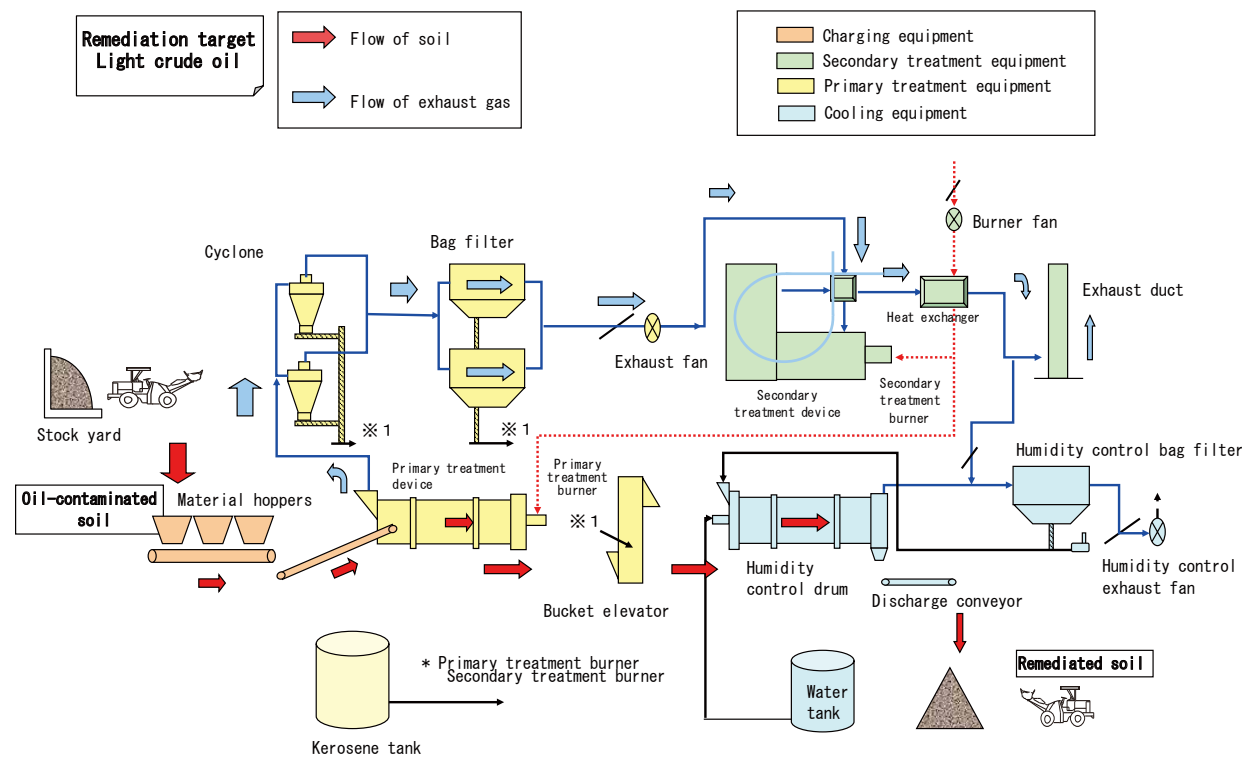

Figure-1 Overall process flow of NTR-Stage I (treatment capacity: 70 t/h)

Photo-4 General view of NTR-Stage I (treatment capacity: 70 t/h)

First, oil-contaminated soil is discharged continuously from the material hopper and fed to the counterflow-type rotary kiln used as the primary treatment device. Here, the oil is desorbed from the oil-contaminated soil by raising the vapor pressure of the oil in the soil is raised to 150 and 250 ℃ to promote vaporization and evaporation.

Concretely, although the boiling point of water is 100 ℃ under atmospheric pressure, laundry dries even when it is not heated to the boiling point of water. Similarly, it is not necessary to heat light crude oils such as gasoline and kerosene to the boiling points of their constituent components. Rather, the rate of evaporation of the oil

constituents in the kiln is governed by their vapor pressure (partial pressure). Based on this fact, the retention time in the rotary kiln is comparatively short, at 5 to 10 minutes, and it is possible to desorb the light crude oils from the soil even at a soil heating temperature lower than the boiling points of the oil constituents, that is, in the temperature range of 150 to 250 ℃.

The heat sources used in the rotary kiln are generally fossil fuels such as city gas, kerosene, A type heavy oil, etc. High-temperature hot blast, which functions as the heat medium, is formed by optimum combustion of these fuels by the burner. Rotary kilns with a metallic structure are equipped with lifters that efficiently rake up, disperse and drop the soil in the kiln. The lifters improve heat exchange by increasing the contact area between the soil and the hot blast.

However, care is necessary, as the moisture content of the soil has a large influence on fuel consumption in this process. That is, so long as water remains in the soil, it is not possible to heat the soil to a temperature higher than 100 ℃ and furthermore, energy is consumed preferentially for evaporation of the water.

Next, the oil components desorbed as gases from the soil, together with the exhaust gas, are introduced into the dust collecting system in the following stage. This exhaust gas includes combustion exhaust gas, infiltrating air and water vapor that evaporates from the soil. In addition, fine soil particles are scattered from the soil which has lost its moisture content as a result of heating and drying, and are discharged from the rotary kiln accompanying the exhaust gas. Since these fine soil particles have an extremely large specific surface area, it is necessary to remember that the oil constituents that were vaporized once will condense again around the soil particles as nuclei as the exhaust gas temperature decreases.

Accordingly, although this system is equipped with dry cyclones and bag filters as a dust collection system, the system is operated in the temperature region where the vapor pressure of the oil will not decrease by raising the temperature of the exhaust gas to near the heat-resistance temperature of the dust collectors. Concretely, because a Teflon heat-resistant filter cloth (tradename: PTFE) is used as the filter cloth of the bag filters, the exhaust gas temperature can be controlled to 200 ℃.

As techniques for controlling the exhaust gas temperature

of the rotary kiln, it is possible to attach or remove the lifters or change the rotational speed (rpm) of the kiln.

Finally, the soil which has been thermally-remediated in the rotary kiln and the fine soil particles captured by the dry cyclone and bag filter system are introduced together into the humidity control drum, where they are sprinkled with water for cooling and as a dust prevention measure, and the soil and fine particles are then discharged from the system in a moist condition.

On the other hand, after filtration by the bag filter, the exhaust gas containing oil components is introduced into the secondary treatment device, where it is decomposed into harmless CO_2 and water by oxidative decomposition at a temperature of around 800 ℃. From the viewpoint of effective utilization of energy, this device is equipped with two heat exchanger units to recover waste heat. The first stage heat exchanger preheats the treated gas, and the second stage preheats the burner combustion air of the primary and secondary treatment devices. Waste heat recovery by these two heat exchangers has achieved an energy saving of more than 30 %. Finally, after waste heat recovery by the heat exchangers, the purified gas is released into the atmosphere through the exhaust gas duct.

3.3 NTR-Stage II

As shown in **Table-1**, the remediation targets of the NTR-Stage II system include the light crude oils which were also the remediation targets of Stage I and heavy oils such as heavy oil, machine oil, etc. These heavy oils are equivalent to oils whose main components are aliphatic hydrocarbons and aromatic hydrocarbons with carbon numbers of C_{10} to C_{23}, and their boiling points are in the range of 150 to 350 ℃. **Photo-5** shows a general view of an NTR-Stage II plant, and **Figure-2** shows its overall process flow.

From this figure, the distinctive feature of the NTR-Stage II system is capture, by the dust collection system, of fine particles with large contents of heavy oils entrained in the exhaust gas from the primary treatment device, and thermal treatment of those particles at 200 to 350 ℃ by a small-scale parallel-type rotary kiln as a tertiary treatment device. In other words, the total system configuration is similar to that of the NTR-Stage I system with addition of the tertiary treatment device. Features of

Photo-5   General view of NTR-Stage II plant

the soil remediation process is presented based on **Figure-2.**

First, when thermal remediation of soils contaminated with light oil and A type heavy oil equivalent was performed with the NTR-Stage I system, failure to achieve the required remediation level was an issue, because a high concentration of oil remained on the fine soil particles captured by a dust collector such as cyclone or bag filter system. Considering the fact that the specific surface area of fine oil particles increases as the particle size becomes smaller and the contaminant oil forms a

steady coating on the soil particle surface, this problem is caused by an increase in the oil content ratio of the particles as the particle size becomes smaller.

From a different viewpoint, it can be understood that the rotary kiln has the characteristics of a classifier, in which the combustion gas functions as the classification medium. The lifters that are installed in the rotary kiln to accelerate heat exchange with the hot blast to rake up, disperse and drop the soil, and heat transfer occurs by contact with the hot blast as it passes through the rotary kiln at a certain speed. In parallel with this, it is thought that light, small-diameter fine soil particles are scattered and entrained in the hot blast.

In this way, the fine soil particles affected by this classifying action become soil particles that are contaminated with condensed, highly concentrated oil due to their large specific surface area. In addition, because the exhaust gas temperature at the outlet of the kiln is limited to 200 ℃, which is the heat-resistance temperature of the filter cloth of the bag filter, oil component that cannot be vaporized and evaporated at this temperature will remain on the surface of the fine soil particles.

Although depending on the type of soil, the amount of fine soil particles scattered from the rotary kiln and entrained in the exhaust gas occupies 10 to 30 % of the total soil, and particularly in the case of the loamy layer found in the Kanto region around Tokyo, which has a high content of silty and clayey soils, this sometimes reaches 60 to 70 %.

Based on these points, one distinctive feature of the NTR-Stage II is a treatment process by which the fine soil particles contaminated with highly concentrated oil, which are captured by the dust collector, are charged into a compact parallel flow-type rotary kiln that serves as a tertiary treatment device, and purification is promoted by reheating the particles to 200 and 350 ℃. At this time, it is also possible to control the exhaust gas temperature in the range of 250 to 400 ℃. Thus, providing a tertiary treatment device that enables purification by vaporization and evaporation of the high boiling point components of the oil that remains on fine soil particles.

To capture the fine soil particles scattered from this tertiary treatment device, a high-temperature dust collector using a ceramic filter material with a heat-resistant temperature of 400 ℃ was adopted. The exhaust gas containing volatile oil components after filtration treatment by the high-temperature dust collector is introduced into the secondary treatment device and detoxified by thermal decomposition together with the exhaust gas from the primary treatment device.

scattered from the rotary kiln and entrained in the exhaust gas at this time, remediation performance is enhanced by controlling the exhaust gas temperature in the range of 250 ℃ to 500 ℃.

In particular, feedback control of the exhaust gas temperature by controlling the burner combustion amount is effective because a parallel flow-type rotary kiln is used. This eliminates the need to control the exhaust gas temperature by attaching/removing lifters or changing the rotational speed of the kiln, as is done in the counterflow-type rotary kilns, in which it is not possible to control the exhaust gas temperature by varying the amount of combustion.

It has already been noted that lifters for dispersing the soil are provided in the rotary kilns with metallic structures used as the primary treatment devices in Stage I to Stage III of the NTR Series. The rotational speed of the rotary kiln is also increased in order to accelerate heat exchange with the hot blast. Because this also shortens the holding time of the soil in the primary treatment device to less than 5 minutes, the NTR-Stage III is provided with a stock bin to promote desorption and aeration of the remaining oil by storing the thermally-treated soil for 30 to 60 minutes to equalize the soil temperature.
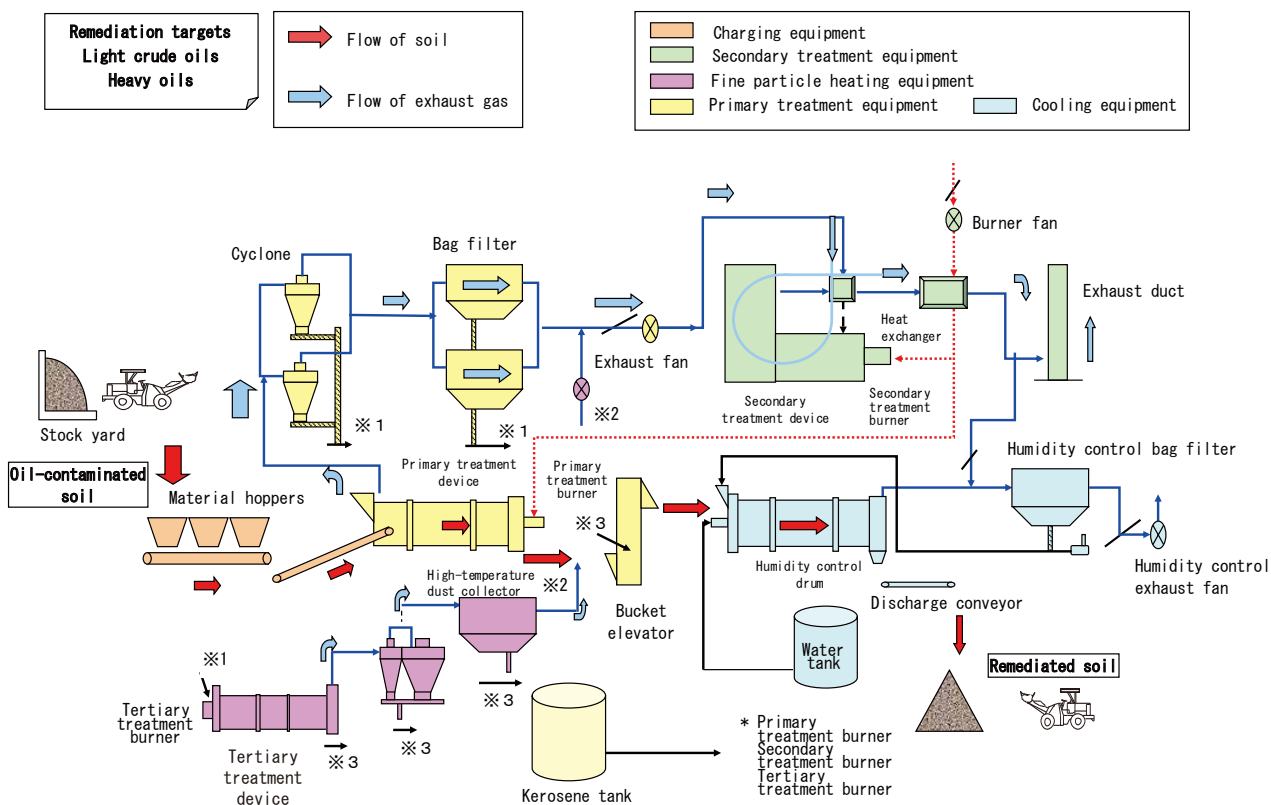
3.4 NTR-Stage III

In addition to the remediation targets of the NTR-Stage II system shown in **Table-1**, the NTR-Stage III system also supports remediation of soil contaminated with heavy oils such as machine oil, tar and pitch, asphalt, etc. and VOC (volatile organic compounds), which are classified as Class 1 designated hazardous substance under Japan's Soil Contamination Countermeasures Law. **Figure-3** shows the overall process flow of NTR-Stage III, and **Photo-6** shows a general view of an NTR-Stage III plant. The following presents an overview of the soil remediation process based on the figure.

First, a parallel flow-type rotary kiln is adopted as the primary treatment device, enabling thermal remediation of contaminated soil in the range of 150 to 400 ℃. Although fine oil particles with adhering heavy oil are
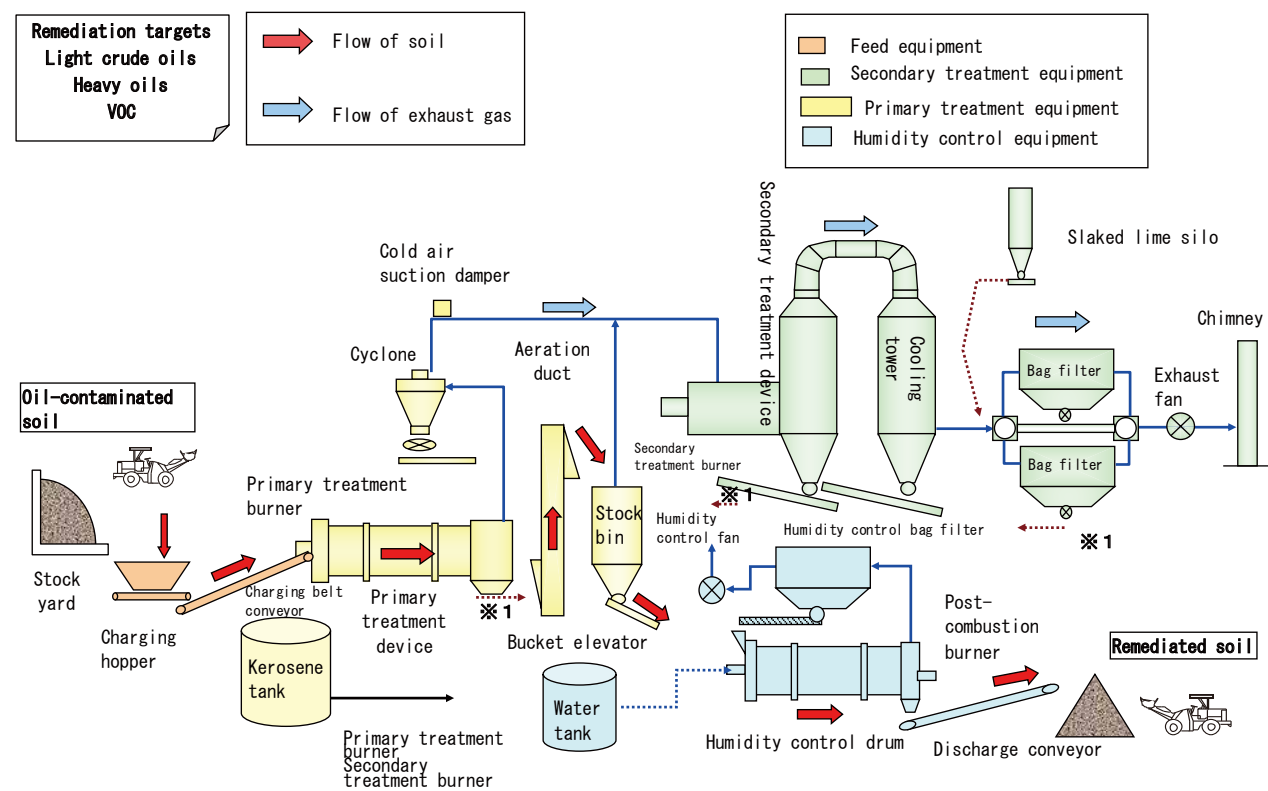

Photo-6 General view of NTR-Stage III plant

Figure-2 Overall process flow of NTR-Stage II

Figure-3 legend (left diagram):

Remediation targets
Light crude oils
Heavy oils
VOC

→ Flow of soil
→ Flow of exhaust gas

Feed equipment
Secondary treatment equipment
Primary treatment equipment
Humidity control equipment

Oil-contaminated soil
Stock yard
Charging hopper
Charging belt conveyor
Kerosene tank
Primary treatment burner
Primary treatment device
Cyclone
Cold air suction damper
Aeration duct
Secondary treatment device
Secondary treatment burner
Bucket elevator
Stock bin
Humidity control fan
Water tank
Humidity control drum
Discharge conveyor
Post-combustion burner
Cooling tower
Slaked lime silo
Bag filter
Exhaust fan
Chimney
Humidity control bag filter
Remediated soil
Primary treatment burner / Secondary treatment burner
※1

Figure-3　Overall process flow of NTR-Stage III [12)]

Figure-4 legend (right diagram):

→ Flow of soil
→ Flow of exhaust gas

Charging equipment
Exhaust gas treatment equipment
Heating equipment
Cooling equipment

Oil-contaminated soil
Stock yard
Material hoppers
Remediation targets
Light crude oils
Heavy oils
VOC
Cyanogen/mercury
Kerosene tank
Heating burner
Primary treatment device
Secondary treatment device
Stock bin
Dry cyclones
Aeration
Bucket elevator
Cooling tower
Slaked lime silo
Activated carbon silo
Option
Exhaust fan
Chimney
Deodorization system
Dust-proofing device bag
Humidity control drum
Discharge belt conveyor
Cooling fan
Water tank
Remediated soil
※Heating burner Deodorizing burner
※1

Figure-4　Overall process flow of NTR-Stage IV

Next, the oil components and VOC desorbed from the soil are detoxified by oxidative decomposition in the secondary treatment device. If soil contaminated with heavy oil is treated at this time, sulfur oxides (SOx), which are subject to emission regulations under the Air Pollution Control Act, will also be precipitated in addition to harmless CO_2 and water, because soil contaminated with heavy oil contains sulfur as well as the hydrocarbons that are its primary components.

In the case of soil contaminated with VOC, hydrogen chloride (HCl), which is also a target of emission regulations, will form in addition to harmless CO_2 and water because the soil contains chlorine components. Since these chlorides can also be a factor in the synthesis of dioxins, the Stage III system is equipped with a cooling tower to prevent resynthesis of dioxins. The cooling tower suppresses resynthesis of dioxins by a *de novo* reaction by rapidly cooling the decomposed purified gas introduced from the secondary treatment device at 800 ℃ to 180 ℃.

The rapidly cooled exhaust gas to 180 ℃ in the cooling tower is then introduced into the bag filter, where fine soil particles are captured and sulfur oxides and hydrogen chloride and other acid gases are neutralized and

removed. The surface of the filter cloth of the bag filter is precoated with slaked lime, and these acid gases react with the slaked lime in the process of capture of the fine soil particles and passage of the exhaust gas through the filter cloth. Together with the fine soil particles, the sulfur oxides are recovered as calcium sulfate (gypsum: $CaSO_4$) and hydrogen chloride is recovered as calcium chloride ($CaCl_2$) and returned to the soil. It should be noted that calcium sulfate and calcium chloride can be returned safely to the soil because both are neutral, nontoxic compounds.

3.5 NTR-Stage IV

In addition to the remediation targets of NTR-Stage III, because the soil heating temperature of the NTR-Stage IV system is increased to 600 ℃, the purification targets of NTR-Stage IV also include Class 2 designated harmful substances such as mercury, cyanogen, etc. and Class 3 designated harmful substances, i.e., agrichemicals and PCBs, as shown in **Table-1**. The overall process flow and general appearance of an NTR-Stage IV plant are shown in **Figure-4** and **Photo-7**, respectively. The following describes the overview of the soil remediation process
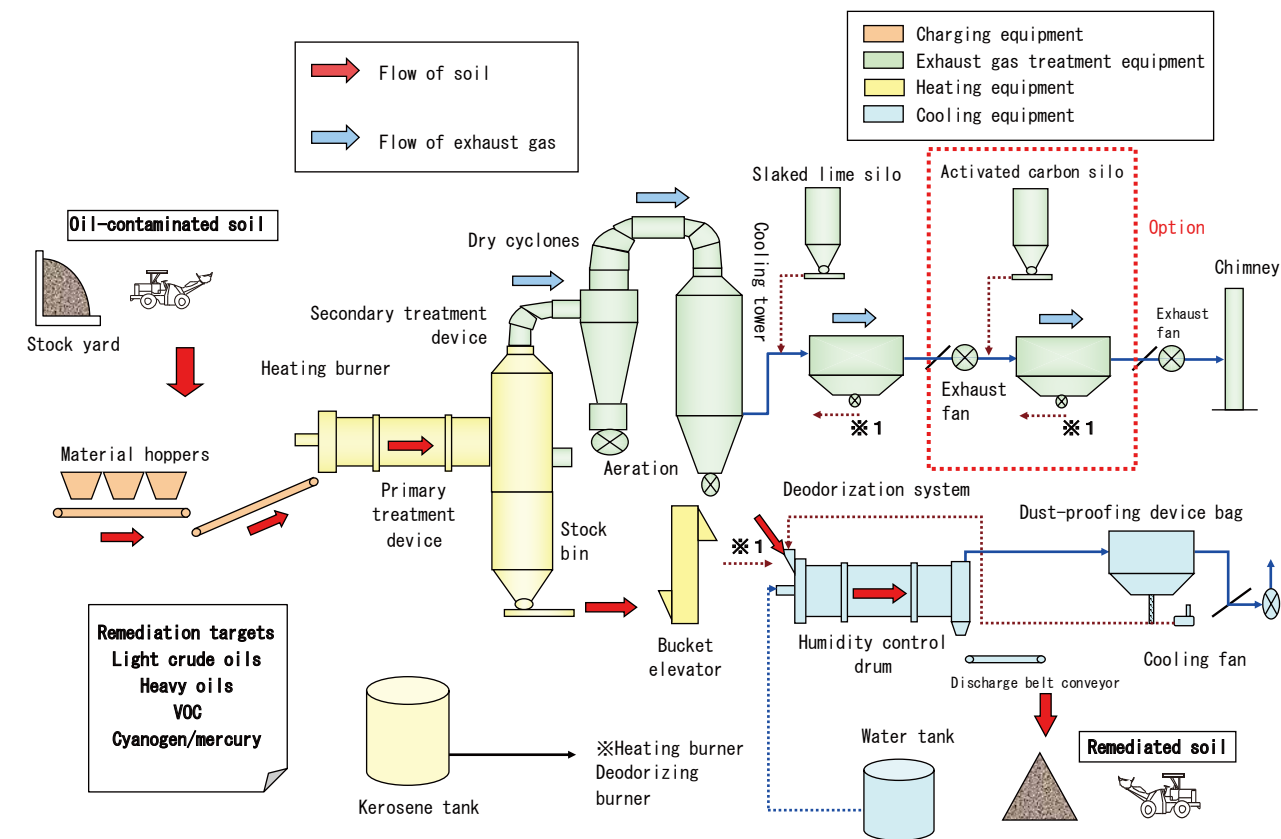
based on **Figure-4**.

As shown in the figure, the NTR-Stage IV system is equipped with a parallel flow-type rotary kiln as the primary treatment device. From the material charging section to the central part of the kiln, it has a metallic structure which is not lined with a refractory castable, and lifters are installed to rake up, disperse and drop the soil. Here, heat exchange efficiency is enhanced by actively dispersing the soil in order to obtain a larger contact area with the hot blast. From the central part of the kiln to the discharge section, the equipment is lined with a refractory castable with an excellent heat storage property responding to the higher soil heating temperature.

Also, as shown in the figure, the primary and secondary treatment devices, and stock bin have a unitary structure and are lined internally with a refractory castable, thereby achieving rationalization and energy saving of the system along with eliminating the need for a high temperature duct extending to the secondary treatment device and a soil conveying system to the stock bin. By combining the exhaust chamber of the primary treatment device and the secondary treatment device, it is also possible to promote gravitational settling of fine soil particles and extend the

Photo-7　General view of NTR-Stage IV plant

retention time of the exhaust gas.

Because the remediation targets of this system include hard-to-decompose agrichemicals, a post-combustion burner is provided in the secondary treatment device, making it possible to achieve a thermal decomposition temperature of up to 1 200 ℃. Since agrichemicals include halogen compounds of fluorine (F), chlorine (Cl), bromine (Br), iodine (I), etc., the acid gases hydrogen fluoride (HF), hydrogen chloride (HCl) and hydrogen bromide (HBr) are formed by oxidative decomposition.
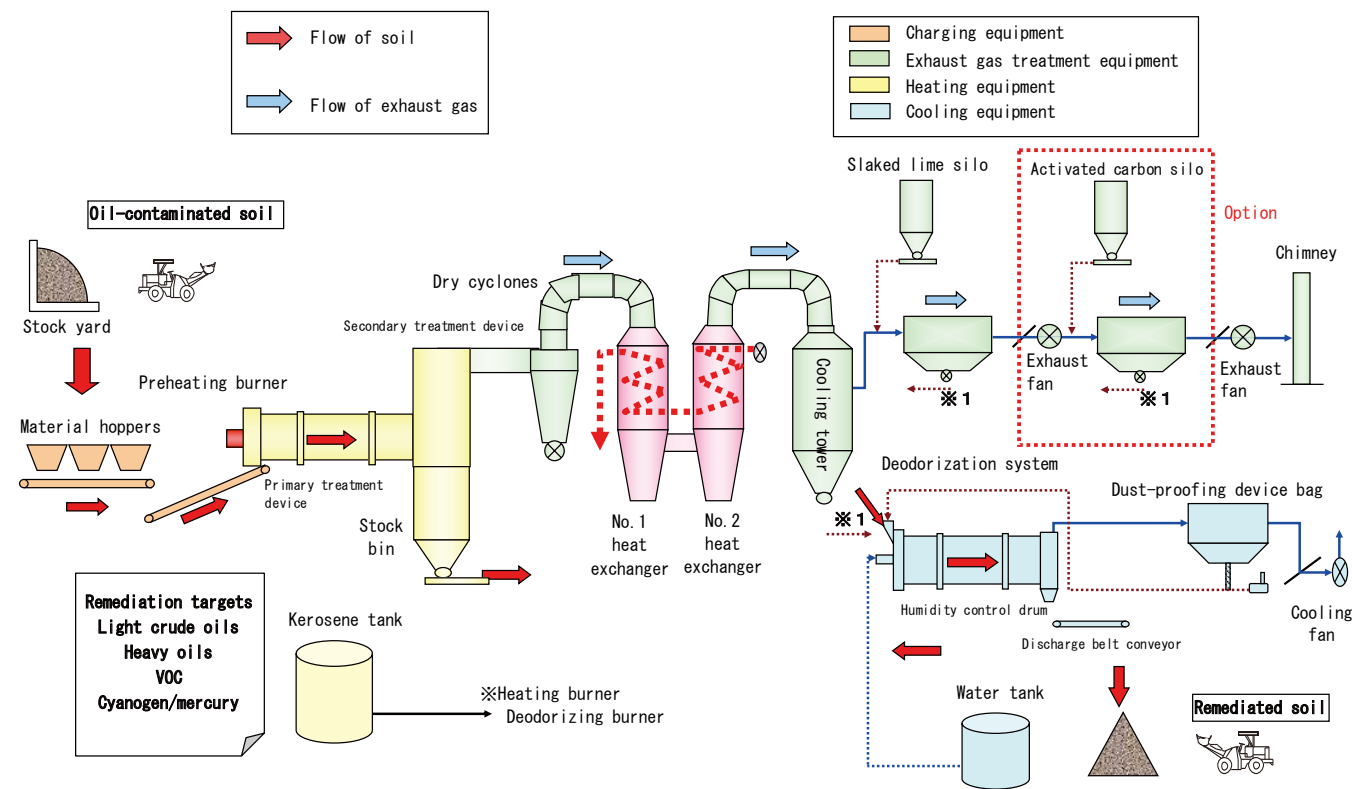
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Figure-5   Overall process flow of NTR-Stage V

As in the NTR-Stage III system, these high-temperature cracked gases are rapidly cooled to 180 ℃ by a cooling tower to suppress the *de novo* synthesis reaction that resynthesizes dioxins, and are discharged into the atmosphere through the exhaust duct after capture of fine soil particles and neutralization of the acid gases with slaked lime in the bag filter. The fine soil particles captured by the bag filter and the calcium fluoride, calcium chloride, calcium bromide and similar compounds formed by neutralizing the acid gases are returned to the soil. The neutralization products of these acid gases are all neutral and nontoxic.

3.6 NTR-Stage V

The NTR-Stage V has the same remediation targets as the NTR-Stage IV, as shown in **Table-1**, but also realizes energy saving by utilizing the waste heat recovered by heat exchangers located downstream from the secondary treatment device, which performs oxidative decomposition of contaminants, to preheat the combustion air of the primary treatment device burner. The overall process flow and a general view of in-house demonstration plant of NTR-Stage V plant are shown in

Photo-8   General view of in-house demonstration plant of NTR-Stage V plant

Figure-5 and **Photo-8**, respectively. The following presents an overview of the soil remediation process, referring to the figure.

From the figure, the entire inner surface of the rotary kiln used as the primary treatment device is lined with a refractory castable with an excellent heat storage property, supporting high temperature thermal remediation of soil, and the soil heating temperature and retention time are controlled by the rotational speed of the rotary kiln. Here, the fact that the soil heating temperature is more

dependent on contact heat transfer and radiant heat transfer from the heat-storing refractory castable than on direct contact with the hot blast is important.

Concretely, although the soil retention time is shortened by increasing the rotational speed of the rotary kiln, the temperature of the soil increases as a result of contact heat transfer from the refractory castable. Therefore, in order to compensate for the short retention time, the soil which has been heated in the rotary kiln is stored temporarily in the stock bin to equalize the soil temperature and promote aeration and oxidative decomposition of the soil.

Fine soil particles scattered from the rotary kiln settle out gravitationally in the secondary treatment device and drop into the stock bin, where they are stored for the specified time together with the soil. Ultra-fine soil particles that do not reach the terminal velocity in the secondary treatment device are introduced into the downstream heat exchangers together with the exhaust gas.

In the heat exchangers, cylindrical tubular elements are used to suppress deposition and growth of clinker and wear due to friction caused by the ultra-fine soil particles entrained in the exhaust gas from the secondary treatment device. In particular, the exhaust gas that entrains the ultra-fine soil particles is introduced in the vertical downward direction from the top of the heat exchanger inside of the tubular elements. This design suppresses clinker deposit and wear by smoothing the flow in the elements. The heat receiving side of the heat exchanger (i.e., outer side of the element tubes) is preheated by introducing clean burner combustion air in order to reduce fuel consumption.

In the flow of the exhaust gas after the heat exchangers, as in the NTR-Stage IV, the exhaust gas is cooled to 180 ℃ by the cooling tower, fine soil particles are captured and acid gases are neutralized in the bag filter, and the exhaust gas is then released into the atmosphere. The fine soil particles and the calcium compounds which are the neutralization products of the acid gases are returned to the soil and humidified by the humidity control drum.

3.7 NTR-Stage VI

As shown in **Table-1**, in the NTR-Stage VI, a preheating kiln was added to the NTR-Stage V system as a soil preheating and drying device to achieve further energy saving. An extremely large energy-saving effect is achieved by introducing the waste heat from the

secondary treatment device into the externally-heated type preheating kiln to reduce the moisture content of the contaminated soil in advance.

As features of the NTR-Stage VI, the remediation performance and energy-saving performance of this system will be described in detail in the next chapter.

4. Newly-Developed Energy-Saving Oil-Contaminated Soil Remediation System (NTR-Stage VI)

4.1 Overview

As mentioned in the previous chapter, the basic principle of thermal remediation systems for oil-contaminated soil is desorption of the target contaminant by increasing its vapor pressure by heating the soil to raise its temperature. Therefore, in the process of heating soil to a temperature of 100 ℃ or more, as long as the soil contains water, the temperature cannot be raised until the soil reaches a dry condition because evaporation of the water consumes energy. Accordingly, if the water content of the soil can be reduced in advance, a large reduction in the energy consumption of the remediation system will be possible.

As mentioned in the previous chapter, the NTR-Stage VI is based on the NTR-Stage V, and a preheating kiln is added as a pre-drying device, as shown in the overall process flowchart in **Figure-6**. As one key feature, the contaminants desorbed from the soil are detoxified by the secondary treatment device, and the high-temperature treatment gas from that unit is introduced into the preheating kiln and effectively utilized to pre-dry the soil.

In addition, surplus heat which is not completely recovered by the preheating kiln is also recovered by a heat exchanger and effectively utilized to preheat the burner combustion air, achieving a large energy-saving effect and CO_2 reduction, as fuel consumption is reduced by half by this cascade use of waste heat. **Photo-9** shows a general view of an NTR-Stage VI plant.

The following describes the details of the remediation process and mechanism of waste heat recovery from the two viewpoints of the flow of the soil and the flow of gases based on **Figure-6.**
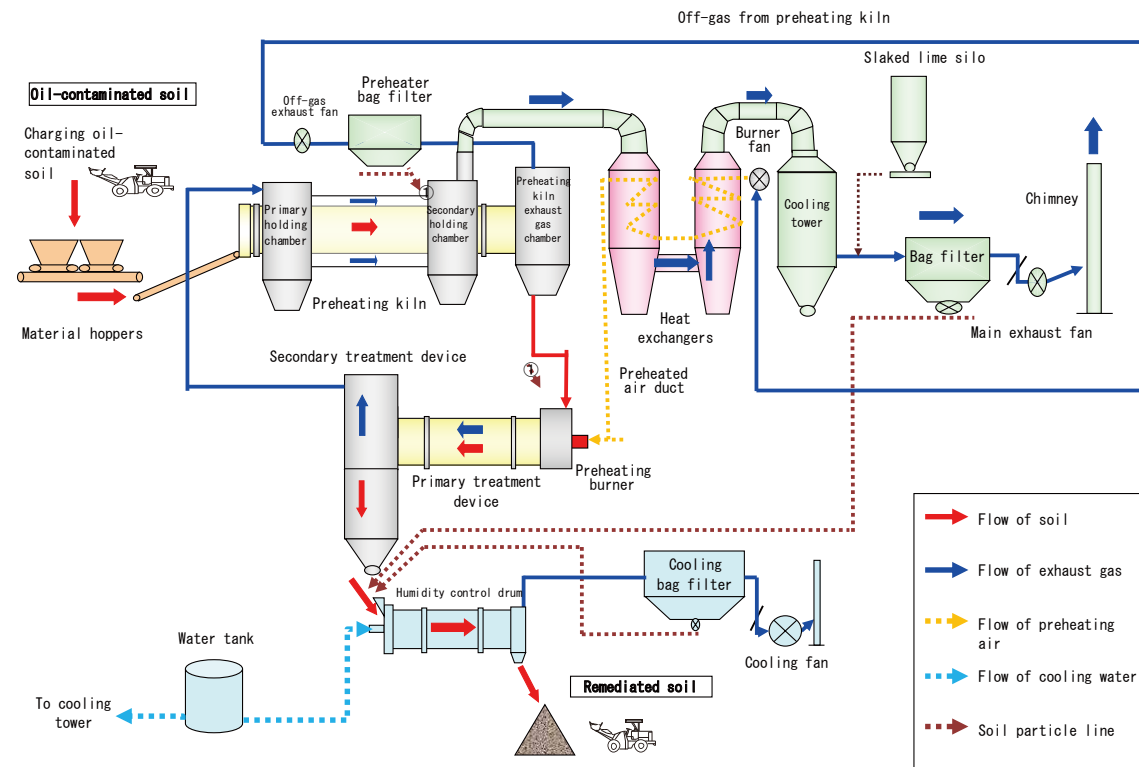
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Figure-6　Overall process flow of NTR-Stage VI

Photo-9　General view of NTR-Stage VI plant

4.2 Viewpoint of Soil Flow

4.2.1 Pre-drying Device (Drying of Soil/Removal of Clods)

As also mentioned in the overview in the previous section, the basic principle of remediation systems for oil-contaminated soil is desorption of oil, which is the contaminant, by heating the soil. For this reason, the moisture content of the soil has a large influence on energy consumption. To mention one example, the thermal energy (latent heat) necessary to evaporate a moisture content of 1 % in soil is equivalent to the thermal energy (sensible heat) needed to heat the soil

30℃, and as long as water exists under atmospheric pressure, the temperature of the soil will not increase to more than 100℃. Thus, in thermal oil-contaminated soil remediation systems, the moisture content of the soil is a large factor in increased energy consumption.

Furthermore, soils are extremely diverse, and soils with a high ratio of silty or clayey soil contain 30 to 50 % moisture. In this case, an enormous amount of energy, exceeding 50 L/t by A type heavy oil conversion, is consumed only as the latent heat of vaporization of water. Thus, how the moisture content can be reduced in the soil pretreatment process is extremely important from the viewpoint of energy consumption.

For example, recovering the waste heat of exhaust gas, which exceeds 850℃ in the secondary treatment device, and utilizing that heat to reduce the moisture content of the soil is very effective for energy saving. The direct heating (internal heating type) kiln and the indirect heating (external heating type) kiln may be mentioned as two means of using waste heat to pre-dry soil.

The basic principle of the former internal heating-type kiln is heat exchange by enabling direct contact between the soil and the hot blast. As a feature of this approach, heat exchange efficiency is enhanced by increasing the

contact area with the hot blast by using lifters provided in the kiln to rake up, uniformly disperse and drop the soil. However, oil components contained in the soil are vaporized and evaporated together with the moisture, and this oil contaminates the hot blast (i.e., exhaust gas) that was specifically detoxified by the secondary treatment device.

The basic principle of the latter external heating-type kiln is indirect heating of the soil in the kiln by raising the temperature of the kiln through contact between the kiln outer surface and high-temperature detoxified exhaust gas used as the hot blast. Therefore, the kiln has a structure that prevents contamination of the hot blast by the volatilized oil components. However, since the heat exchange capacity depends on the contact surface between the kiln and the soil, a larger scale kiln will undeniably be required in comparison with the internal heating type.

For this reason, an external heating-type kiln with a metallic structure was adopted as the preheating kiln used as a pre-drying device in this system.

Concretely, the preheating kiln itself is heated by flowing detoxified high-temperature exhaust gas as a heat carrier (heat medium) on the outside part of the preheating kiln, while soil is charged inside the kiln, and heat transfer is limited to the thermal energy that passes through the wall surface of the preheating kiln. This enables heat transfer of only thermal energy to the soil, without contamination of the detoxified exhaust gas. Based on **Figure-6**, the following describes this process in detail following the flow of the soil. **Photo-10** shows a general view of the preheating kiln.

As a precondition, first, this process assumes pretreatment of the oil-contaminated soil i.e., the remediation target, to a condition in which stable feeding by a continuous-type belt feeder is possible. Specifically, the properties of the soil are improved by removing any large foreign matter with heavy equipment and performing quicklime addition or other appropriate treatment. The soil is charged continuously into the preheating kiln via the material hoppers, and is heated by the kiln wall surface by contact heat transfer. Because the preheating kiln is heated to 400 to 600℃ by the high-temperature exhaust gas, far-infrared and infrared radiation are radiated toward the soil in the kiln. In

particular, because far-infrared radiation reaches the interior of the material being heated and the amount of heat transfer is proportional to the fourth power of the temperature difference, radiant heat transfer is controlling factor when the kiln is in the high-temperature region.

The former type of heat transfer, contact heat transfer from the preheating kiln wall surface to the soil, depends on the contact area between the soil and the kiln and the heat transfer coefficient. Therefore, it is important to increase the soil retention time and expand the heating area. Since the latter radiant heat transfer also depends on the apparent area of the retained soil surface and thermal emissivity, increasing the amount of retained soil is effective, and the amount of radiant heat transfer is greater than that of contact heat transfer.

In addition, as shown in **Photo-11**, lifters are installed in the preheating kiln to promote soaking and exhausting of moisture by turning over the soil.

After completion of the drying process, which requires a retention time of about 1-hour, large foreign matter is removed from the dried soil by a trommel installed at the outlet of the preheating kiln, as shown in **Photo-12**, and the soil is transferred to the primary treatment device, which is the next heating process.

Photo-10　General view of preheating kiln of NTR-Stage VI

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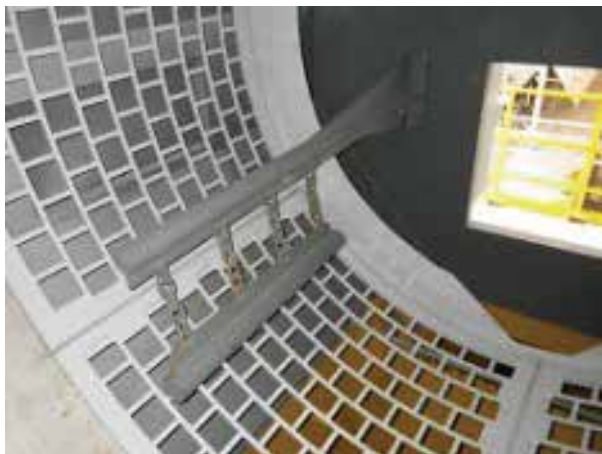Photo-11　Interior of preheating kiln of NTR-Stage VI

Photo-12　Trommel part of preheating kiln of NTR-Stage VI

Photo-13　General view of primary treatment device and stock bin of NTR-Stage VI

Photo-14　General view of secondary treatment device and stock bin of NTR-Stage VI

Photo-15　Interior of primary treatment device of NTR-Stage VI

4.2.2 Primary Treatment Device (Heating of Soil)

The rotary kiln which belongs to the primary treatment device has a heat-storing structure in which the interior of the kiln is lined with a refractory castable. This heat-storing refractory castable makes it possible to heat contaminated soil to 400 to 600℃. **Photo-13** and **Photo-14** show general views of the secondary treatment device and stock bin, and **Photo-15** shows the condition of the refractory castable lining inside the rotary kiln.

First, the refractory castable on the inner surface of the rotary kiln is heated by a high-temperature hot blast generated by burner combustion and stores this heat. Heat storage by the castable at this time involves both heat transfer by direct contact with the hot blast, and heat transfer by radiation from the hot blast. Because burner combustion is fundamentally diffusive combustion which

forms a red-hot flame, the flame radiates infrared and far-infrared radiation, causing heat storage in the refractory castable. In particular, because contact heat transfer is proportional to the temperature difference, while radiant heat transfer is proportional to the fourth power of the temperature difference, radiant heat transfer becomes controlling factor as the temperature difference increases.

Similarly, heat also is transferred from the refractory castable, which has stored heat at a high temperature, to the low-temperature soil by a combination of contact heat transfer and radiant heat transfer. Although the temperature of the refractory castable decreases by an amount corresponding to heat transfer by direct contact with the soil that accumulates in the kiln bottom, the castable is returned to a high temperature by the heat received from the hot blast when the rotation of the rotary kiln returns the castable to the region where it is not in contact the soil. In addition, heat is also transferred from the refractory castable toward the soil by radiant heat transfer. In this way, the refractory castable plays the role of a heat carrier which transfers the heat received from the high-temperature hot blast to the low-temperature soil. Moreover, although the proportion of direct heat transfer from the hot blast to the soil is not particularly large, its contribution must also be considered.

Next, the exhaust gas temperature of the secondary treatment device is controlled to the setting temperature of 850℃ by feedback control of burner combustion. For example, if heat transfer to the soil through the refractory castable becomes excessive, the temperature of the castable will decrease, and together with this, the temperature of the secondary treatment device will also decrease, so burner combustion is increased corresponding to that difference.

Control of the heating temperature of the soil is performed by adjusting the rotational speed of the rotary kiln. For instance, under a constant soil feeding rate, the amount of soil retention (mass) will decrease if the speed of the kiln is increased, and at the same time, the contact area between the soil and the refractory castable also decrease, but this is because the rate of decrease is larger in the case of retention (mass). That is, retention (mass) per unit of contact area of the soil is changed by changing the

rotational speed of the rotary kiln, and also has a negative correlation with the rotational speed of the kiln.

Based on this, if the speed of the rotary kiln is increased, the temperature of the soil will increase due to the decrease in the amount (mass) of retained soil per unit of contact area, and conversely, if the speed is reduced, the temperature of the soil will decrease because the amount of retained soil increases.

In this connection, in the relationship between the rotational speed of the kiln and retention time, the soil retention time generally decreases when the speed of the kiln is increased. Conversely, when the speed of the kiln is decreased, the retention time becomes longer. There is also a tradeoff relationship between the soil heating temperature and retention time: If the speed of the kiln is increased to raise the temperature of the soil, retention time will decrease, and conversely, if the speed of the kiln is decreased to lower the soil temperature, retention time will increase. The degree of that influence (sensitivity) is larger for rotational speed than for retention time.

While the soil heating temperature and retention time are the main factors in soil remediation performance, based on the points discussed above, the main purpose in the rotary kiln is the control of the soil heating temperature, and retention time is assigned to the downstream stock bin.

4.2.3 Stock Bin (Soaking and Aeration of Soil)

In the stock bin, aeration and oxidative decomposition of the remaining oil are promoted by soaking of soil, that is, by holding the heated soil until it reaches a uniform temperature. **Figure-7** shows the image of the interior of the stock bin. Although also mentioned in the preceding section, a tradeoff relationship exists between the heating temperature and the retention time of soil in the rotary kiln, in that the retention time in the kiln decreases as soil is heated to higher temperatures. Moreover, since it is a basic principle of heating that heat transfer proceeds from the outer surface of soil particles to the center, the oil and VOC adhering to the surface of soil particles reach a high temperature and are easily vaporized and evaporated, but as the size of the soil particles increases, the temperature in the center is low, making it difficult for residual oil impregnated in the interior to escape. In addition, it is

also thought that the heating temperature of soil changes due to uneven distribution of the soil particle size and changes in the soil feed rate.

Considering these points, a uniform soil temperature can be achieved by storing the soil heated in the primary treatment device in a stock bin for a specified time so that large, low-temperature particles can receive heat from high-temperature small particles, as shown in **Figure-7**. Soaking also promotes vaporization and evaporation of the low-temperature residual oil impregnated in the interior of soil particles. In addition, storing soil heated to a high temperature in this manner causes a chimney effect due to the temperature difference between the soil and the outside air, and the air that enters the stock bin through crevices in the discharge gate at the bin bottom accelerates the progress of purification by oxidative decomposition of the residual oil in the soil.

Figure-8 shows the relationship between the heating time of soil (rotary kiln retention time + stock bin retention time) and the oil concentration of the soil. The soil heating temperature at this time is 200℃, the exhaust gas temperature is 300℃ and the remediation standard is 1 000 ppm. **Figure-9** shows the relationship between the soil heating time and the oil residual ratio, which is the nondimensionalized value of the oil concentration. **Figure-10** shows the relationship between the heating time of the soil normalized for the oil concentration before remediation and remediation efficiency.

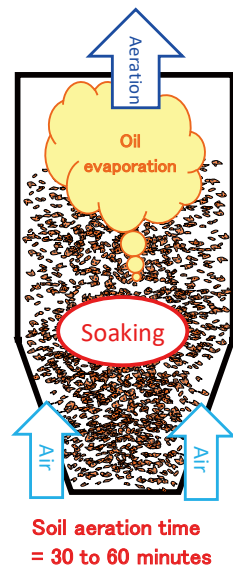From these figures, focusing on 5 minutes, which is

Figure-7 Image of interior of stock bin [12]

equivalent to the heating time in the rotary kiln, even though the oil concentration of the kerosene equivalent, which is a light crude oil, did not clear the remediation standard of 1 000 ppm, the concentration decreased to 1440 ppm. Based on this value, the oil residual ratio decreased to 15.7 %, showing remediation efficiency of 84.3 %. On the other hand, the oil concentration of the machine oil equivalent, a heavy oil, decreased to 4840ppm, and based on this, the oil residual ratio was 32.0 % and remediation efficiency was limited to 68.0 %.

Next, looking at the results for 30 minutes, that is, adding the storage time in the stock bin to the heating time in the rotary kiln, the oil concentration of the kerosene equivalent decreased to 92 ppm, which satisfies the remediation standard of 1 000 ppm by a wide margin. Based on this, the oil residual ratio decreased to 1.0 %, thus achieving a remediation efficiency of 99.0%. The oil concentration equivalent to machine oil decreased to 670 ppm, which also satisfies the remediation standard, and from this, the oil residual ratio decreased to 4.5 %, achieving remediation efficiency of 95.5 %. From these results, it can be surmised that the residual oil content was reduced as a result of the oxidative decomposition reaction and the action of aeration, irrespective of the low soil heating temperature of 200℃.

Based on the results described above, storage of the heated soil in the stock bin for the specified time is considered to be an extremely effective means of achieving soaking of soil and promoting aeration and oxidative decomposition of residual oil. Moreover, since the remediation level is not increased by indiscriminately raising the soil heating temperature, but is heightened by storage time in the stock bin, this technology is also considered to be extremely effective for energy saving and reduction of CO₂ emissions.

4.2.4 Humidity Control Drum (Humidity Control of Soil)

In the humidity control drum shown in **Figure-6**, the soil is returned to its original moist condition by sprinkling water for cooling and dust prevention. The cooling effect in this process is enhanced by using lifters installed inside the humidity control drum to rake up, disperse and drop the soil.
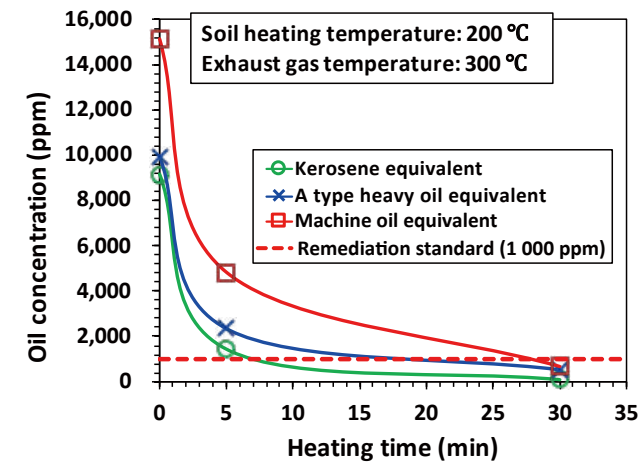

Figure-8 Relationship between soil heating time and oil concentration [12]
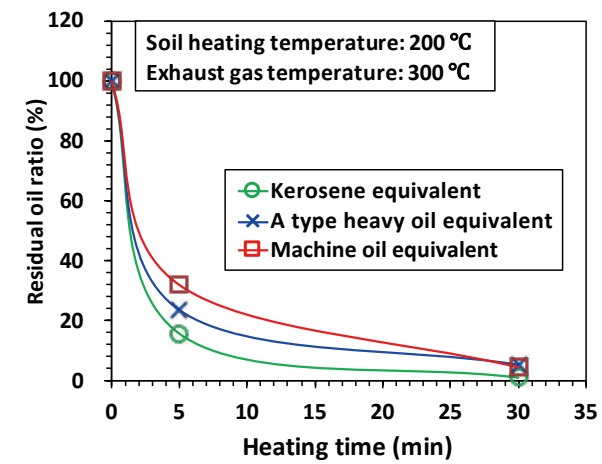

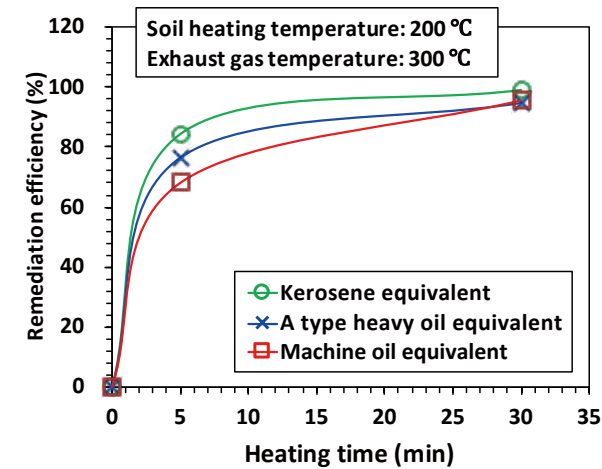Figure-9 Relationship between soil heating time and oil residual ratio [12]

Figure-10 Relationship between soil heating time and remediation efficiency [12]

In order to remove the steam generated by sprinkling water, a large volume of outside air is introduced into the humidity control drum and is then suctioned to a bag filter. A counterflow design in which the outside air and the soil flow in opposite directions is used in the humidity control drum. This makes it possible to control the temperature of the treated gas at the bag filter inlet by controlling the amount of sprinkling water and amount of outside air introduced in the humidity control drum. The temperature of the treated gas is controlled in the range of 120℃ or higher in order to avoid low-temperature corrosion and condensation in the bag filter, to not more than 200℃, which is the heat-resistance temperature of the filter cloth.

The fine soil particles entrained in the steam and outside air in this process are captured and recovered by the bag filter, returned again to the humidity control drum, and then discharged together with the soil as remediated soil. A storage hopper equipped with a discharge gate is installed at the discharge outlet of the humidity control drum, making it possible to control the timing of transfer of soil to the heavy equipment used to transport the soil.

4.3 Viewpoint of Gas Flow

The following traces the flow of gas from the upstream side based on the process flowchart of the NTR-Stage VI system shown in **Figure-6**.

4.3.1 Heat-Receiving Side of Pre-drying Device (Interior of Preheating Kiln)

In addition to the explanation of the preheating kiln from the viewpoint of the soil flow in section 4.2.1, this section also mentions on its features from the viewpoint of the gas flow.

As shown in **Figure-6**, the starting point of the flow of gas in this system is the soil charging section of the preheating kiln. From here, the air necessary for combustion by the burner of the primary treatment device is sucked into the system. Because the oil-contaminated soil is retained and heated in the preheating kiln, the moisture and volatile components contained in the soil are vaporized and evaporated, and then mix with the combustion air. In addition, because the soil is stirred by the lifters attached to the inner side of

the preheating kiln, as shown in **Photo-11**, the dried fine soil particles scatter and are entrained in this combustion air. The temperature of the combustion air at this time rises to the range of 100 to 200℃, which is the temperature of the dry soil on the discharge side of the preheating kiln.

Next, after the combustion air has passed through the preheating kiln, fine soil particles are captured by filtration by the preheater bag filter, and the dedusted air is sucked into the heat-receiving side of the heat exchanger. The fine soil particles captured and recovered by the preheater bag filter are transported to the primary treatment device together with the soil, and soil remediation is performed by thermal decomposition.

Because the details of the heat exchanger will be described in section 4.3.6 below, only an outline will be presented here. The heat exchanger is a 2-pass type which is divided into a low-temperature side and a high-temperature side, and cylindrical tubular elements are adopted, as shown in **Photo-16**. The exhaust gas on the heat radiation side of the heat exchanger, which contains entrained fine soil particles (exhaust gas from the secondary treatment device which has passed through the heat-radiation side of the preheating kiln) is introduced into the inner side of these tubular elements, while the combustion air on the heat-receiving side (combustion air filtered by the bag filter of the preheating kiln) is introduced on the outer side of the elements, which have a complex bundled structure. The combustion air is heated to 300 to 500℃ by absorbing waste heat and supplied to the preheated air burner of the primary treatment device, which is explained in the following section.

4.3.2 Preheated Air Burner of Primary Treatment Device

Combustion air containing steam and volatile components of oil is preheated to 300 to 500℃ in the heat exchanger and supplied to the preheated air burner of the primary treatment device shown in **Photo-17**. Here, countermeasures for thermal NOx are necessary because this burner raises the temperature of the preheated air to 500℃ as an energy-saving measure.

Therefore, an Exhaust Gas Recirculation system (hereinafter, EGR system) has been introduced in the NTR-Stage VI. The EGR system reduces the oxygen concentration in the combustion air by recirculating part of the combustion exhaust gas as combustion air. This suppresses high-temperature regions that appear locally by suppressing fast burning, thereby reducing precipitation of thermal NOx.

Concretely, part of the combustion exhaust gas is extracted from the chimney, introduced into the combustion air which has passed through the preheating kiln bag filter, and mixed. In the mixing ratio of EGR, the oxygen concentration in the combustion air is controlled arbitrarily in the range of 21 to 17 % by way of an oxygen concentration meter installed in the combustion air duct to this burner. The setting value of this oxygen concentration is changed according to the regulatory value for NOx emissions based on full-time monitoring of the NOx concentration in the exhaust gas by a continuous NOx concentration meter on the chimney. Although the NOx value will decrease if the oxygen concentration is reduced, the concentration of carbon monoxide (CO) will increase. In particular, an excessive increase in the CO

concentration will result in incomplete combustion, and thus will have the opposite effect from energy-saving.

In other words, because there is a tradeoff relationship between the NOx concentration and the CO concentration, the optimum oxygen concentration is set considering the values of both.

Photo-18 shows the interior of the mixing chamber of the preheated air burner. As can be seen in the photo, a divided flame is produced by installing a radial arrangement of fuel gas injection nozzles at six points. This design suppresses precipitation of thermal NOx by preventing the formation of local high-temperature hot spots in the flame.

Thus, the NTR-Stage VI responds to emission regulations on nitrogen oxides by the effects of the EGR system and divided flame. It may be noted that the fuel used with this burner is 13A city gas, which contains methane as its main component and has the minimum CO_2 emission of any fossil fuel.

On the other hand, because the preheated air burner supports combustion air with temperatures from room temperature to 500℃, the air volume varies by up to 2.7 times. Since the combustion range is TDR = 1 : 5 and the excess air ratio varies from 1 to 2, the amount of air passing through the burner (air velocity) also varies from 1 to 27 times, and because the amount of air (air velocity) is proportional to the square of pressure, the static pressure of combustion changes from 1 to 729 times.

In particular, an optimum value of the static pressure of combustion exists in combustion of 13A city gas. If the value is low, the combustion air will stall and mixing with the fuel will deteriorate, resulting in a dull flame shape. Conversely, if the static pressure of combustion is increased, mixing with the fuel gas will be accelerated and a sharp flame shape will form. Furthermore, if the static pressure of combustion exceeds a certain value, combustion vibration will occur due to abnormal combustion (detonation). If this occurs, phenomena such as vibration of the burner unit or mechanical vibration due to resonance with the rotary kiln may also occur, resulting in damage to the equipment.

The factors of the mixing velocity of the combustion air in the burner and the static pressure of combustion are governed by the size of the diffuser (flame stabilizer). In ordinary burners, the size of the diffuser is adjusted so that the amount of air necessary for the maximum amount of combustion under the highest combustion air temperature condition can be supplied. This means that the combustion air will stall and the condition of combustion will deteriorate under conditions of a small combustion amount with room temperature air.

To solve this problem, a diffuser slide mechanism is provided, as shown in **Figure-11**. This mechanism can continuously change the clearance (section area) formed between the mixing chamber and the outer circumference of the diffuser by continuously sliding the diffuser forward and backward.

In case the combustion air temperature is high and the combustion amount is large, the mechanism prevents abnormal combustion by sliding the diffuser forward to increase the clearance (cross-sectional area) so that the static pressure of combustion (mixing rate) does not increase above a certain level. Conversely, when the combustion air temperature is low and the combustion amount is small, the mechanism prevents stalling of the combustion air (sluggish combustion) by sliding the diffuser backward, thereby decreasing the clearance (cross-sectional area) so that the static pressure of combustion (mixing rate) does not fall below a certain level.
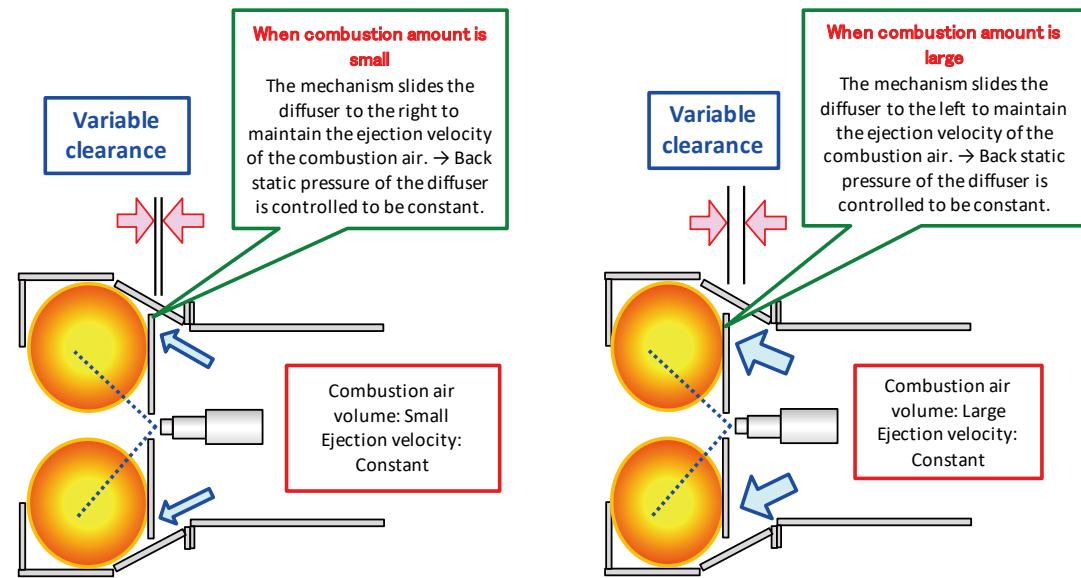

Photo-16　Heat-receiving gas inlet on side of heat exchanger of NTR-Stage VI

Photo-17　Preheated air burner of primary treatment device of NTR-Stage VI

Photo-18　Interior of burner of primary treatment device of NTR-Stage VI

Figure-11   Diffuser slide mechanism of primary treatment device burner of NTR-Stage VI

4.3.3 Primary Treatment Device (Rotary Kiln)

As mentioned in the detailed description of the primary treatment device (rotary kiln) from the viewpoint of the soil flow in section 4.2.2, the hot blast generated by combustion of the preheated air burner heats the refractory castable of the inner lining of the rotary kiln, and the soil is heated from the refractory castable, which has stored high-temperature heat, through the heat transfer mechanisms of contact and radiation.

The burner combustion gas contains the steam and volatile components of oil generated in the preheating kiln and the EGR gas. Therefore, when a large amount hot blast is generated by burner combustion and passes through the rotary kiln, it picks up fine particles of the soil in the kiln and is introduced into the secondary treatment device together with those particles.

In the rotary kiln, static pressure control that can reduce the internal pressure in the kiln slightly from the outside pressure has been introduced to reduce the amount of air entering from the rotary seal. This is contributing to stabilization of burner combustion by controlling the static pressure in the rotary kiln to a constant value at all times, even when the amount of burner combustion or the soil feed rate changes.

4.3.4 Secondary Treatment Device (Secondary Combustion Device)

The thermal decomposition treatment temperature in the secondary treatment device is controlled by the amount of burner combustion by feedback control. In order to detoxify exhaust gas that may possibly contain dioxins, this secondary treatment device conforms to the equipment standards applicable to waste incinerators, which are required to maintain a retention time of 2 seconds at a temperature of 850℃ or higher. The soil heating temperature in the rotary kiln used as the primary treatment device is controlled by the rotational speed of the kiln, as mentioned in section 4.2.2.

In **Figure-12**, ,the vertical axis on the left shows the relationship between the concentration of total hydrocarbons (hereinafter, THC), which are the volatile components of oil, and the decomposition temperature of the secondary treatment device, and the axis on the right shows the relationship between the THC decomposition efficiency and decomposition temperature of the secondary treatment device. In all cases, the retention time is 2 seconds.

As can be seen in this figure, from the point when the secondary treatment temperature exceeds 550 °C until it reaches 650 °C, the THC concentration decreases rapidly and the decomposition efficiency on the right axis increases rapidly. From 650 °C to 800 °C, the THC concentration gradually approaches 0 ppm, while the decomposition efficiency gradually approaches 100 %. These results support the fact that the decomposition temperature is an extremely important factor in oxidative decomposition of the volatile components of oil into carbon dioxide and water. Where THC decomposition efficiency is concerned, factors other than the decomposition temperature, such as retention time, oxygen concentration, stirring speed, etc. are conceivable.

In the fine soil particles scattered from the rotary kiln, hard-to-decompose organic compounds and other substances adhere to the particles and some of them impregnate to the particle interior. Among those particles, comparatively large fine soil particles are dropped into the stock bin, together with soil heated to 600 °C, by gravitational settling, and are remediated by oxidative decomposition and the aeration action during soaking in the bin. On the other hand, comparatively small fine soil particles, which are entrained in the exhaust gas, are remediated by exposure to a temperature of 850 °C or higher for 2 s in the secondary treatment device. In both cases, the organic compounds desorbed from the soil in the stock bin and the scattered fine soil particles are finally detoxified by oxidative decomposition into carbon dioxide and water.

VOC, etc. including halogen elements react with water vapor in the oxidative decomposition process and form acid gases such as hydrogen fluoride (HF), hydrogen chloride (HCl), hydrogen bromide (HBr) and others. These acid gases are neutralized by slaked lime $(Ca(OH)_2)$ precoated on the filter cloth of the later-stage bag filter, and are then recovered and returned to the soil as neutral, harmless calcium fluoride (CaF_2), calcium chloride $(CaCl_2)$ and calcium bromide $(CaBr_2)$.

Oil-contaminated soil contains mineral oil and organic matter of animal and plant origin. These substances include sulfur compounds and nitrogen compounds, and precipitate sulfur oxides and nitrogen oxides under the oxidative decomposition process in the secondary treatment device.

The former sulfur oxides (SOx) are neutralized (lime-gypsum desulfurization method) in the high-temperature secondary treatment device by quicklime (CaO), which is added in the oil-contaminated soil pretreatment stage, and slaked lime $(Ca(OH)_2)$ added to the soil on the charging side of the preheating kiln, and are captured and recovered as harmless gypsum $(CaSO_4\cdot 2H_2O)$ by the bag filter in the final process and then returned to the soil. Gypsum is also obtained by
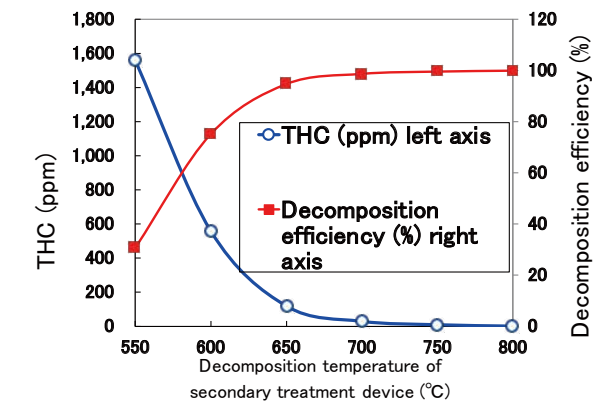

Figure-12   Effects of decomposition temperature in secondary treatment device on THC and decomposition efficiency

neutralization by the slaked lime precoated on filter cloth of the bag filter, as mentioned above. The amounts of slaked lime addition to the bag filter and soil are determined based on the results of continuous measurements of the SOx concentration in the exhaust gas by a SOx concentration meter installed on the chimney.

Regarding the latter nitrogen oxides (NOx), since NOx originates from the soil, the NOx reduction effects of the divided flame of the burner and EGR cannot be expected. Therefore, NOx is reduced by adding urea water $(CO(NH_2)_2 + H_2O)$ to the combustion gas in the secondary treatment device. Concretely, ammonia (NH_3) is synthesized by hydrolysis of urea, and is reduced to N_2 and H_2O by reaction with NOx. The reactions at this time are shown in Eq. (1) and (2).

$$CH_4N_2O + H_2O \rightarrow NH_3 + CO_2 \qquad (1)$$
$$2NH_3 + 2NO + 1/2\ O_2 \rightarrow 2N_2 + 3H_2O \ (2)$$
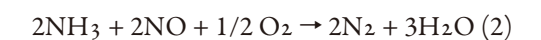
Although discussed in detail in section 4.3.2, the amount of urea water addition is determined based on continuous measurements of the NOx concentration in the exhaust gas with a NOx concentration meter on the chimney and a total evaluation with the amount of thermal NOx precipitation of the primary treatment device.

4.3.5 Heat-Release Side of Pre-drying Device (Outside of Preheating Kiln)

Although mentioned in section 4.2.1 in detail, the high-temperature treated gas with a temperature of 850℃ or higher detoxified by the secondary treatment device is introduced into the heat-release side (outside) of the preheating kiln, which is a pre-drying device. The kiln is a parallel flow type, as the flow direction of the hot blast on the outside and flow direction of the contaminated soil inside the device are the same. This design suppresses temperature increases in the steel shell of the preheating kiln.

Because the preheating kiln consists of austenitic heat-resistant steel, the temperature of the steel shell is controlled to 600℃ or less. Concretely, the surface temperature of the preheating kiln is continuously monitored with radiation thermometer, and if the temperature exceeds the preset temperature, the high-temperature treated gas is bypassed to prevent excessive temperature rise in the steel shell.

The fine soil particles entrained in the hot blast gas settle out in various parts of the preheating kiln. These fine soil particles are recovered by a screw conveyor and returned to the humidity control drum as remediated soil.

4.3.6 Heat Exchanger

The heat exchanger is a 2-pass type which is divided into a low-temperature side and high-temperature side, as shown in **Figure-6**, and is installed so that the flow of gas is vertical in both passes. The burner combustion air on the heat-receiving side is introduced from the low-temperature side and reaches the high-temperature side, while the exhaust gas on the heat-receiving side is introduced from the high-temperature side and travels to the low-temperature side. Thus, the heat exchanger is a counterflow type that can achieve high temperature effectiveness over the entire temperature range.

To reduce the effects of clinker formation and growth by fine soil particles entrained in the exhaust gas and the accompanying wear, the cylindrical tubular elements shown in **Photo-19** are adopted. Naturally, the heat-releasing side gas, which contains fine soil particles, is passed through the inner side of the elements, where there are few obstacles to its flow. Finally, the entrained fine soil

particles are settled out and deposited in a hopper provided at the bottom of the heat exchanger, and are then recovered by a screw conveyor, transported to the humidity control drum, and finally returned to the soil.

The burner combustion air on the other side, i.e., the heat-receiving side, is introduced from the outer side of the elements, as shown in **Photo-16**. In the elements on the heat-receiving sides, turbulence is generated by providing baffle plates at several points to improve the uniformity of the gas temperature and increase heat exchange efficiency, and expansions are installed at the bottom of the cylindrical tubes to absorb thermal expansion and contraction of the elements.

An exhaust gas bypass circuit is also provided to prevent excessive temperature rise in the steel shells of the elements and suppress rises in the preheated air temperature on the heat-receiving side, and to control the temperature of the exhaust gas on the heat-releasing side so that it does not fall below 400℃, because the *de novo* synthesis reaction, that is, the dioxin resynthesis reaction, proceeds in the temperature range of 350 to 300℃.

Photo-19 Top of heat exchanger
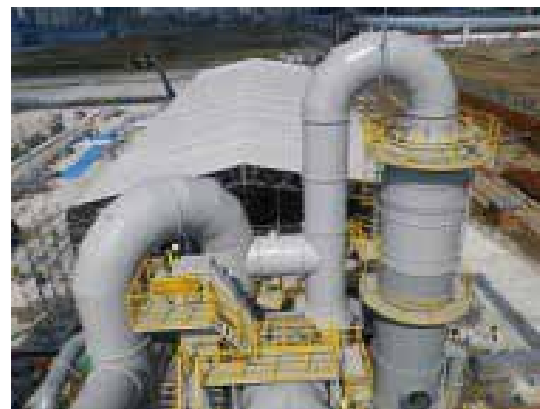**(heat-release side gas is introduced into the tubes)**

Photo-20 (From the left) Preheating kiln,
heat exchanger and cooling tower

4.3.7 Cooling Tower (Cooling of Exhaust Gas)

From the left, **Photo-20** shows that the exhaust gas duct of the preheating kiln is connected to the top of the heat exchanger. The bypass duct, which horizontally connects the two vertical ducts is visible in the center, and the cooling tower is on the right.

The cooling tower has the function of rapid cooling the exhaust gas to avoid the above-mentioned resynthesis of dioxins. The principle of exhaust gas cooling is cooling by the latent heat of vaporization of water by spraying water directly on the high-temperature gas. The retention time of the exhaust gas in the cooling tower is set at 4 seconds or longer, and the gas is rapidly cooled instantaneously from around 500℃ to 180℃. A two-fluid spray nozzle is used to achieve this rapid cooling effect, as it is necessary to reduce the average particle diameter of the water spray to 50 microns or less.

The cooling tower is a downdraft type, in which the exhaust gas flows vertically from the top to the bottom, and is a perfect evaporation-type device in which water spraying is performed from the top and the water is completely evaporated by the time it reaches the bottom of the cooling tower. As a result, drain is not generated in the tower bottom. A heater is installed in the tapered part at the bottom of the tower to cope with the drain generated in the initial stage of operation.

Although fine soil particles settle out in the bottom of the cooling tower due to the downdraft design, the particles are blown upward by sucking in outside air from the tower bottom, and are then carried to the bag filter. These basic concepts of the cooling tower are common to the systems from NTR-Stage III to NTR-Stage VI.

4.3.8 Bag Filter (Neutralization and Capture of Acid Gases)

After the exhaust gas has been cooled to 180℃ by the cooling tower, the bag filter captures and recovers the fine soil particles in the exhaust gas, and the resulting dust concentration conforms to regulatory values. Together with this, the recovered fine soil particles are transported to the humidity control drum and returned to the soil.

Although mentioned in previous sections, the surface of the filter cloth is precoated with slaked lime to neutralize the sulfur oxides and other acid gases formed by oxidative decomposition of oil components in the secondary

treatment device. When the exhaust gas passes through the filter cloth, a neutralization reaction with the slaked lime occurs. Although the types of acid gases depend on the contaminants, gases such as sulfur oxides, hydrogen fluoride, hydrogen chloride, hydrogen bromide, etc. are assumed. These acid gases react with the slaked lime and are recovered as calcium sulfate ($CaSO_4$), calcium fluoride (CaF_2), calcium chloride ($CaCl_2$) and calcium bromide ($CaBr_2$). Because these neutralization products are harmless, they recovered together with the fine soil particles and returned to the humidity control drum as soil.

Deliquescent compounds such as calcium chloride ($CaCl_2$) are also formed in the neutralization reaction with slaked lime in the bag filter. Therefore, temperature is controlled by supplying power to an electrothermal heater in the bag dust-collection hopper to prevent clogging of the filter cloth in case operation of the remediation plant is stopped temporarily. Power is not supplied while the plant is in operation because the temperature of the treated gas is 180℃.

4.4 Energy-Saving Performance

Table-2 shows the specifications of the NTR-Stage IV, NTR-Stage V and NTR-Stage VI systems and a comparison of their energy-saving performance. According to a trial calculation of the difference in the annual fuel costs assuming a contaminated soil treatment capacity of 25 t/h, moisture content of 30 %, heating temperature of 600 °C, exhaust gas purification temperature of 850 °C, annual operation time of 7 200 h and unit cost of 13A city gas fuel of ¥100/Nm³, the fuel cost difference due to the energy-saving effect of Stage VI is ¥519.84 million in comparison with Stage IV and ¥326.88 million in comparison with Stage V.

In addition, assuming the unit CO_2 emission of 13A city gas is 2.36 kg/CO_2/Nm³, the CO_2 reduction effect of Stage VI is 12 268 t/y in comparison with Stage IV and 7714 t/y in comparison with Stage V.

From the relative comparison of the exhaust gas volumes in the table, the exhaust gas volume of Stage VI is reduced to 53 % in comparison with Stage IV. Because the equipment cost of exhaust gas treatment facilities from the secondary treatment device and cooling tower to the dust collection system makes up 70 % of the total cost of

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thermal soil remediation equipment, reduction of the exhaust gas volume has a large impact on the cost reduction effect. This can also result in a relative reduction in electric power costs.

These results support the fact that the moisture content of soil has a large effect on the treatment capacity, fuel consumption, equipment costs, running costs and CO_2 emissions of thermal soil remediation plants. Reducing the moisture content of the soil in advance by utilizing the waste heat of the treated gas is considered to be an extremely effective method with excellent economic rationality, including equipment costs and operating costs.

Table-2　Performance comparison of soil remediation systems

Comparison item	Stage IV	Stage V	Stage VI
Treatment capacity (t/h)	25	←	←
Moisture content (%)	30	←	←
Soil heating temperature (°C)	600	←	←
Exhaust gas purification temperature (°C)	850	←	←
Fuel type (-)	13A	←	←
Lower heating value (Kcal/Nm³)	9,970	←	←
Annual operating time (h)	7,200	←	←
Fuel unit cost (Nm³/¥)	100	←	←
Preheater heating area (m²)	0	←	80
Soil preheating temperature (°C)	10	10	75
Soil residual moisture (%)	30	←	20
Heat exchanger heating area (m²)	0	250	200
Preheated air temperature (°C)	15	480	341
Preheated air volume (Nm³/min)	440	335	273
Fuel consumption (Nm³/h)	1,898	1,444	1,176
Fuel consumption per ton of soil (Nm³/t)	75.9	57.8	47.0
Exhaust gas temperature (°C)	180	←	←
Exhaust gas volume (Nm³/min)	917	673	488
Exhaust gas volume ac (Nm³/min)	1,521	1,117	811
Cooling tower sprinkling rate (L/h)	13.076	6,400	2,947
Relative comparison of fuel consumption (%)	100.0	76.1	62.0
Relative comparison of exhaust gas volume (%)	100.0	73.4	53.3
Relative comparison of sprinkling water amount (%)	100.0	48.9	22.5
Difference of annual fuel cost (¥10 000)	0	-32,688	-51,984
Annual CO_2 reduction (t/y)	0	-7,714	-12,268

5. Future Technological Prospects for Oil-Contaminated Soil Remediation Systems

The following discusses the feasibility of thermal technology for soil contaminated with heavy metals, which are classified as Class 2 designated hazardous substances in Japan, and technologies for energy saving and reduction of CO_2 emissions.

5.1 Thermal Remediation Technology for Heavy Metal-Contaminated Soil

The possibility of thermal-based detoxification of compounds classified as Class 1 (VOC) and Class 3 (agrichemicals) designated hazardous substances under the Soil Contamination Countermeasures Law and its effectiveness have become clear. However, because almost all Class 2 designated hazardous substances are heavy metals and their elements themselves are classified as hazardous substances, it is not possible to detoxify these elements by thermal decomposition. Although technologies that convert these substances to harmless elements by nuclear fission, nuclear fusion or the like may be applied practically in the future, this approach will not be discussed here, as the timeframe is far from the present.

As a practical method for remediation of soil contaminated with heavy metals, the current mainstream technique is washing. In this method, soil clods are purified by extracting particles of fine soils such as silt and clayey soil which have comparatively large surface areas from soil clods by classification action. Since heavy metals adhere to the surface of soil particles, they are concentrated in the fine soil particles extracted from soil clods. Effective utilization of these fine soil particles after separate special treatment or final disposal is also necessary. Concretely, the methods of effective utilization as a raw material for cement by mixing a certain amount of these particles in cement kilns and insoluble treatment followed by disposal in a controlled disposal site are current conditions.

On the other hand, one practical method for removing heavy metals from soil by a thermal technology is volatilization and removal by addition of an alkali metal or chloride to the contaminated soil to lower its melting point or boiling point. In particular, the chloride volatilization method, which is an application of

this technology, is used as a refining technology for nonferrous metals such as copper, lead and zinc, and for removal of heavy metals from incineration ash and other types of waste. In the latter case, the incineration ash is converted to molten slag, and is then effectively utilized in civil engineering materials.

Thus, effective utilization of fine soil particles classified by the washing method as civil engineering materials after removal of heaving metals by this volatilization technique is expected.

In another practical example, the Great East Japan Earthquake of 2011 caused a manmade disaster in the form of radioactive contamination when the reactors at the Fukushima No. 1 Nuclear Power Plant melted down and spread radioactive cesium-137 over a wide surrounding area. Radiation-contaminated earthquake disaster debris was incinerated to reduce its volume, and incineration ash with a high level of radioactivity was provisionally stored at temporary storage sites. Similarly, surface soil with a high level of radioactive contamination was also removed from farmlands, homes, etc. and provisionally stored at temporary storage sites. The number of these temporary storage sites reached 1 118 locations, and the total volume area rose to 16.5 million m³.

The above-mentioned chloride volatilization method is one effective method for separating and concentrating the cesium-137 from this incineration ash and contaminated soil and reducing its volume. In this method, calcium chloride is added to the contaminated soil or incineration ash and forms cesium-137 chloride, which is then cooled and condensed, and captured and recovered by a bag filter.

Thermal remediation of soil contaminated with heavy metals in this manner is a purification process which is performed in a temperature range exceeding 1 000 °C. Since this process forms clinker and molten slag, it will also be necessary to simultaneously promote the development of applications for these byproducts as civil engineering materials.

5.2 Reduction of CO_2 by Deep Cultivation of Energy-Saving Technologies

Because oil-contaminated soil remediation plants using the basic principle of heating consume large amounts of fossil fuels at a heat source, deep cultivation of energy-saving technologies is extremely important from

the viewpoints of CO_2 reduction and reduction of fuel costs. Among those technologies, recovery of the waste heat of high-temperature exhaust gases and use in soil preheating is a very effective energy-saving technology because the moisture content of the soil has an extremely large impact on energy consumption. In the NTR-Stage VI system, it was possible to achieve a certain level of energy saving by recovering the waste heat of the exhaust gas and pre-drying the soil in an indirect heating-type preheating kiln.

In the current system, the treated soil is backfilled after sprinkling water with water while still hot following thermal remediation. This is done as a dust prevention measure and in order to return the soil to its original moist condition, but to promote further energy saving, recovery and effective utilization of the sensible heat of this heated soil is also conceivable. For example, the sensible heat of soil heated to 600 °C is comparable to the thermal energy that can reduce the moisture content by approximately 10 %. Therefore, additional effective utilization of this heat in pre-drying of soils with high moisture contents is expected.

6. Conclusion

In response to the Soil Contamination Countermeasures Law enacted in 2003, countermeasures for potential soil contamination have been taken and soil contamination in Japan has decreased, if only gradually. Under the revision of the Soil Contamination Countermeasures Law of April 1, 2018, when altering land with an area exceeding 900 m², the history of the site must be investigated, and if the possibility of soil contamination is suspected based on that history, a boring survey must be carried out to determine whether the ground is actually contaminated or not.

When contamination exceeding the environmental standards for soil is detected, a report to this effect must be submitted to the administrative authorities, and if prevention of the spread of contamination is a concern, the owner is also obligated to take countermeasures to prevent its spread. The contaminated area is also designated as a "designated area," and in this case, alteration is difficult and the appraised value of the land will also decrease.

Based on this, the usual countermeasures when soil contamination is discovered are excavation and removal or in-situ remediation of the contaminated soil. In case of excavation and removal, the soil must also be transported to a designated plant specified by law.

On the other hand, looking at overseas countries, it is thought that soil contamination similar to that in Japan is also a potential problem in the emerging economies, which are continuing to enjoy rapid economic growth. In particular, in the background of rapid economic growth under governments that prioritize economic growth at the expense of the environment, as in Japan in the past, legal regulations have been applied to air pollution, water quality, noise and vibration, but measures for soil pollution are inadequate.

Based on our company's drying-heating technologies and deodorization technologies for asphalt plants, beginning in 1999, Nikko successively developed a series of remediation systems for oil-contaminated soil, which has a higher degree of difficulty than asphalt. While our track record does not yet extend to remediation of soil contaminated with PCB or dioxins, which are extremely difficult to decompose, we are steadily enhancing our technical capabilities to enable treatment of those contaminants. In the future as well, we will ceaselessly devote ourselves to the study of remediation technologies, and will unstintingly commit management resources to building a new system for remediation of soil contamination and reduction of environmental loads through our corporate activities.

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