Current status of NOx emission treatment in Marine Diesel Engine

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Abstract. Nitrogen oxide (NOx) of marine diesel engine is a great danger to environment, which is one of the world’s recognized atmospheric pollutants. In this paper, on the basis of describing the international emission regulations and the NOx generating route, we have introduced the emission control technology and the development tendency of the pretreatment, in-machine purification and after-treatment. It can provide some guidance for China’s research and development.

1. Introduction
Diesel engines are widely used in ships for their high power, good economy and high reliability. More than 95% of ships in the world use diesel engines as power [1, 2], but development of the diesel engine is restricted by seriously exhaust emissions. NOx from diesel engine exhaust is an important source of air pollution. IMO[3] (International Maritime Organisation) statistics show that diesel powered ships in the world emit about 10 million tons of NOx into the atmosphere every year, and the pollution caused by NOx to human activities and atmospheric environment has attracted extensive attention at home and abroad [4]. Such regulations are more stringent than IMO regulations.

To cope with the environmental problems, the IMO in September 1997 by the convention on the 73/78 pollution prevention (MARPOL convention) supplement IV, which established a ship NOx emission regulations was put into effect in May 2005. In 2011, the Tier II emission regulation was implemented, which stipulated that the NOx emission of Marine diesel engines was 25% lower than the limit of Tier I emission. In early 2016, emissions Control area (ECAs) began to implement Tier III laws and regulations, the EGAs (in the Baltic sea, the north sea, etc.), the ship must conform to the Tier III Emission standards; When a ship sails outside the ECAs, it shall at least meet Tier II standards. Tier III is 80% lower than the limit of NOx emission of Tier I. In addition, Marine diesel engines must comply with local emissions regulations for coastal areas when passing through certain routes or ports. Such regulations are generally more stringent than IMO emissions regulations.
2. The formation of nitrogen oxides

Due to high compression ratio, short combustion time, uneven mixture of diesel, rough combustion process and high excess air coefficient, the diesel engine is prone to form a high-temperature and oxygen-rich environment in the cylinder, which results in the formation of a large number of NOx.

There are two kinds of NOx in diesel engine: NO and NO\textsubscript{2}, in which the content of NO is 90% ~ 95%. The emission types and generation process of NOx in diesel engines are as follows [4]:

1) thermal NO. Thermal NO is the main source of NO produced by actual diesel engine combustion. At high temperature, N\textsubscript{2} reacts with O\textsubscript{2} to produce NO. Thermal NO production is closely related to temperature, O\textsubscript{2} concentration and high temperature residence time. Controlling the high-temperature residence time in the cylinder can effectively reduce NOx emissions [6].
2) fast NO. This type of reaction is very fast, so it is called fast NO. In the combustion process of diesel engines, the proportion of fast NO is also very small, generally less than 5%. During the combustion of a diesel engine, a large number of free radicals are generated on the front surface of the HC flame, and the major areas of rapid NO formation occur in the incomplete combustion zone with carbon amino groups.

3) fuel NO. Fuel NO is produced by the oxidation of the element N in the fuel. The content of N in diesel is very high, and that in heavy oil is up to 0.2% ~ 0.5% [4]. The reaction mechanism of fuel NO is very complicated, and the reaction process is not sensitive to temperature, which is closely related to fuel air ratio. When the fuel air ratio $\Phi > 1$, the air increase, fuel type NO increase; When the fuel air ratio $\Phi < 1$, continue to increase the air, type fuel NO fell sharply.

3. NOx emission reduction technology for Marine diesel engines

3.1 Fuel pretreatment

Pretreatment refers to the pretreatment of fuel or air before it enters the cylinder to reduce NOx produced during combustion in the cylinder. Preprocessing mainly includes the following contents [7,8]:

1) limit aromatic hydrocarbon content. The aromatics in the fuel increase the PM and NOx in the diesel engine exhaust and lead to an increase in the pahs in the exhaust, which are more toxic. Controlling the content of aromatics in diesel fuel, especially the content of pahs, can effectively reduce the concentration of NOx.

2) adjust the cetane content. Marine diesel has a lower cetane number (usually less than 40), which causes the diesel to burn longer in the cylinder. The amount of fuel injected is large, leading to prolonged premixed combustion and a sharp increase in the pressure and temperature of the gas in the cylinder after the fuel fire, which promote the formation of thermal NOx. Therefore, according to the fuel stroke, reasonable adjustment of the content of cetane in the fuel can make the combustion process more complete and reduce the concentration of NOx of the incomplete combustion products.

3) reduce sulfur content. The lower the sulfur content in diesel, the lower the SO2 emission in its gas. SO2 can lead to sulfur poisoning of NOx catalyst, and reducing sulfur content in diesel is an effective method to improve the activity of NOx catalyst.

4) dual-fuel engine. Techniques such as limiting aromatics, adjusting hexadecane and reducing sulphur are expensive and complex and are not suitable for most ships. Dual-fuel engines can solve this problem. Dual-fuel engines use natural gas as the main fuel and diesel as the ignition fuel, which can reduce NOx emissions and reduce fuel consumption [9]. Compared with conventional diesel engines, the amount of ignition diesel is very small, so the NOx produced by diesel is very small [10]. The flame propagation rate of natural gas is low, the temperature in the cylinder is low, and the mixture of natural gas and air is very thin, so it can't reach the combustion environment with high temperature and rich oxygen at the same time in both high and low load, and NOx emission is reduced [11]. Under the low load condition of diesel engine, controlling the concentration of gas-air mixture and increasing the amount of diesel can improve the combustion of natural gas, but the combustion temperature will increase, which will increase NOx emission. At this point, the EGR technology is added and increase the EGR rate, the working medium in the cylinder is diluted, the oxygen concentration decreases, the heat capacity of the mixture increases, the combustion temperature decreases, and the NOx emission decreases [12].

3.2 Internal Purification technology

3.2.1 Combustion chamber optimization

NOx is formed in diesel engines under conditions of oxygen-rich, high temperature and long temperature duration. The design of combustion chamber is the key factor to influence these
parameters. The design of combustor shape can improve the quality of mixer and the emission performance of diesel engine.

Homogeneous gas-filled compression ignition HCCI[13], premixed diesel combustion [14], premixed or partially hybrid compression ignition PCCI[15], reaction controlled compression ignition low-temperature combustion RCCI[16], jet controlled compression ignition JCCI[17] can control NOx emissions.

3.2.2 Fuel injection system optimization
Factors such as timing of diesel injection, fuel injection pressure, fuel injection rate and injection diameter have effects on NOx generation and emission. Delayed fuel injection, increased injection pressure, multiple injection and optimized fuel injection system can effectively reduce NOx emissions. The widely used application is high pressure common rail fuel injection technology. High pressure common rail fuel injection technology refers to the fuel provided by the oil pump with a certain pressure and stored in the common rail track, shared by each cylinder, and kept the same injection pressure in full operation state, according to control different working conditions of the diesel engine control injection timing, fuel injection volume, reduce NOx emissions. At present, the maximum injection pressure of the high pressure common rail fuel injection technology in the laboratory can reach 300MPa, and the actual production can reach 250MPa[18,19].

The high-pressure common rail electronically controlled fuel injection system can be adjusted by the previous mechanism or program to achieve the injection timing ahead and behind. When timing lags, the combustion process lags, peak combustion temperature decreases, and NOx emissions decrease. Not only that, Injection lag can also reduce the duration of high temperature and reduce NOx emissions. However, improper use of this technology will greatly increase PM and HC emissions in diesel engine exhaust, and fuel economy and engine power performance will also deteriorate [20].

The high pressure common rail fuel injection technology is equipped with piezoelectric ejectors to achieve high precision and multiple injection at short intervals in a shorter response time. It can not only reduce NOx emissions, but also reduce PM emissions. High interference ejectors, and each set of nozzle holes which formed by the intersection of two sub-jets, can control the diameter of nozzle holes, the diameter of the total outlet, the intersection Angle, etc., which make the control of fuel spray characteristics more flexible and accurate, and can adapt to different types of combustion chamber shapes. The higher disturbance ejector can effectively promote oil and gas mixing, control combustion rate, reduce NOx emissions, and achieve higher fuel economy [21].

3.2.3 Charge adjustment
The miller cycle is adopted in the high-power diesel engine, and the inlet valve is closed in advance, which can shorten the inlet stroke and compression stroke, reduce the effective compression ratio, and reduce the temperature of end compression, thus reducing NOx emission. However, due to the shortened opening time of the inlet valve, in order to keep the air-fuel ratio consistent with the original engine, a supercharger with a higher pressure ratio is usually required to compensate for the lost intake charge.

Studies have shown that the use of different miller timing, combined with a variety of injection strategies, can effectively reduce NOx emissions. However, the fuel injection strategy needs to be re-adjusted for different valve timing [22].

3.2.4 Humidifying technology
Diesel combustion in oxygen-rich conditions releases a large amount of heat, which accumulates to produce high temperature, and produces a large amount of NOx in the region. By injecting liquid water into the combustion chamber, the water evaporates and absorbs some of the heat, reducing the temperature in the cylinder and the temperature of the combustion flame, thus inhibiting the formation of NOx. Humidifying techniques include FWE, HAM and DWI.
FWE refers to adding appropriate amount of emulsifier and water to diesel fuel to form emulsified diesel oil. During the combustion process, the water phase in the water-in-oil particles rapidly evaporates at high temperature, and the fuel particles break up and disperse immediately, and are fully mixed with the air. Water evaporates at high temperatures and absorbs a lot of heat, reducing the maximum combustion temperature in the cylinder and thus reducing NOx emissions. The oil emulsification method is simple to operate and can easily reduce the emission of NOx by 20% [23]. However, emulsifier costs are high, consumption is too large, emulsified oil is not easy to use on fire, and the emulsification effect is good only for heavy oil, while it is difficult to emulsify diesel and light diesel, which all limit the use of oil emulsification.

HAM refers to mix atomized water and air in the inlet humidifier, then evaporates after passing through the intake of the diesel engine and into the supercharger. Saturated air is formed to lower the temperature of the mixture, which then enters the cylinder and is mixed evenly with diesel oil before burning. Compared with FWE, the NOx emission reduction effect of HAM is more obvious due to the increase of water, and the influence on the durability and reliability of diesel engines is small [24]. Combined use of FWE and HAM can reduce NOx emissions by more than 50% [4].

DWI refers to the use of the original fuel injection system or the second injection system directly into the combustion cylinder at high pressure (20MPa~50MPa) to reduce NOx emissions. The water-oil ratio is greatly increased and NOx emission can be reduced by 50% ~ 70%. It can also switch between "with water" and "without water" modes. However, DWI needs to make major structural changes to diesel engines, which increases costs, and shortens nozzle life, increase fuel consumption and PM emissions. Moreover, to avoid white and black smoke at low loads, water volume must be reduced or the sprinkler system turned off [25].

3.2.5 Selective non-catalytic reduction technology (SNCR)
Selective non-catalytic reduction (SNCR) refers to the process in which the reducing agent (usually urea) is injected into the cylinder without catalyst and rapidly decomposed into ammonia at high temperature (about 850°C~1100°C), reducing NOx in the flue gas to N2 and water vapor. SNCR technology does not need catalysts, and the control cost is low. However, improper control will lead to a large amount of ammonia escape, which will react with SO3 in the exhaust gas, NH4HSO4 and (NH4)2SO4, leading to corrosion and blockage of the exhaust pipe [26].

3.3 After-treatment technology
Internal Purification technology is for the reconstruction of the existing ship diesel engine, which results in the higher initial investment cost, the complex technology, and only to a certain extent reduction of NOx. A separate internal control technology can only meet the IMO Tier II emission standards, in order to meet more stringent emission standards, after-treatment technology must be employed to diesel engine.

3.3.1 Exhaust gas recirculation technology (EGR)
Exhaust gas recycling technology is to mix some diesel engine exhaust with air through the intake pipe, and then enter the diesel engine cylinder for combustion with fuel, thus reducing the oxygen content in the combustion chamber, reducing the combustion temperature in the cylinder and reducing the combustion rate [27], thus reducing the generation of NOx.

The EGR system consists of an internal EGR and an external EGR, and the exhaust of the internal EGR circulates in the cylinder. Common methods used to achieve internal EGR include exhaust gas residual method (eliminating valve overlap Angle), exhaust gas back suction method, etc. The exhaust utilization pipe of the external EGR will introduce the exhaust gas through the cooler from the exhaust pipe to the intake manifold, mix with the air, and enter the cylinder for combustion. Compared with internal EGR, external EGR can not only accurately control the EGR rate, but also reduce the oxygen concentration in the intake, increase the content of inert gas in the intake, and cool the exhaust gas to reduce NOx emission [28].
3.3.2 Selective catalytic reduction (SCR)
SCR (Selective Catalyst Reduction) refers to the selective reduction of NOx to N2 by a reducing agent (NH₃, HC, CO) in the presence of a Catalyst. The selective catalytic reduction technology has high efficiency, up to 90%.

In industry, urea solution is decomposed into NH₃ at high temperature, and the equation is as follows [29,30].

\[
\text{CO(NH}_2\text{)}_2(\text{aq}) \rightarrow \text{CO(NH}_2\text{)}_2(\text{s}) + \text{H}_2\text{O(g)}
\]

\[
\text{CO(NH}_2\text{)}_2(\text{s}) \rightarrow \text{NH}_3(\text{g}) + \text{HNCO(g)}
\]

\[
\text{HNCO(g)} \rightarrow \text{NH}_3(\text{g}) + \text{CO}_2(\text{g})
\]

Under the action of the catalyst, NH₃ reacts with NOx to form pollution-free N₂ and H₂O, and the equation is as follows.

\[
4\text{NH}_3 + 4\text{NO} + \text{O}_2 \rightarrow 4\text{N}_2 + 6\text{H}_2\text{O (standard reaction)}
\]

\[
4\text{NH}_3 + 2\text{NO}_2 + \text{O}_2 \rightarrow 3\text{N}_2 + 6\text{H}_2\text{O}
\]

\[
2\text{NH}_3 + \text{NO}_2 + \text{NO} \rightarrow \text{N}_2 + 3\text{H}_2\text{O (rapid reaction)}
\]

In the absence of oxygen, the equation is:

\[
4\text{NH}_3 + 6\text{NO} \rightarrow 5\text{N}_2 + 6\text{H}_2\text{O}
\]

\[
4\text{NH}_3 + 6\text{NO}_2 \rightarrow 7\text{N}_2 + 12\text{H}_2\text{O}
\]

The catalyst is the core of the whole reaction. Catalysts used in SCR reaction of diesel engine mainly include metal oxide catalyst, noble metal catalyst, carbon-based catalyst and zeolite catalyst. At present, metal catalysts are widely used in Marine diesel engine SCR system. V₂O₅-WO₃-TiO₂ is a mature catalyst, which has been successfully used on the ship. V₂O₅ is the active material, TiO₂ is the carrier and WO₃ is the auxiliary agent, which can improve the thermal stability of the catalyst. Temperature is very important for the SCR reaction. First, the reaction temperature of SCR is usually between 200°C and 600°C, and the optimum temperature of V₂O₅-WO₃-TiO₂ catalyst is between 300°C and 400°C. Secondly, low temperature is not conducive to the pyrolysis reaction of urea aqueous solution, and a long time of high temperature will lead to the sintering of the catalyst, and when the exhaust temperature is too high, NH₃ and NO will be burned. In addition, the concentration and atomization degree of NH₃ will also directly affect the efficiency of SCR reaction. When NH₃ concentration is too high, unreacted NH₃ will be discharged into the atmosphere and cause pollution.

SCR technology, which first appeared in coal-fired power plants and oil refineries, has been applied to ships since the 1990s and has developed into the most common NOx removal method on ships. There are two main SCR systems for Marine diesel engines, high pressure system and low pressure system [31]. The exhaust gas temperature before the supercharger is about 300°C~450°C, and the pressure can be as high as 4bar; the exhaust temperature after the supercharger is about 210°C~260°C, and the pressure is about 0.03bar. In contrast, the SCR is placed in front of the supercharger as a high voltage SCR system, and the SCR is placed behind the supercharger as a low voltage SCR system. The catalytic reduction temperature of the exhaust gas needs to be greater than 280°C, so that the high-pressure SCR system can meet the requirements. The high-temperature environment of the high-pressure SCR system will also reduce the production of hydrogen sulfate and ammonia and reduce secondary pollution. However, the high-pressure SCR system is arranged near the main engine, with large vibration and high requirements for the materials of pipelines and accessories, and the equipment is bulky and the installation is not flexible. The low-pressure SCR system is arranged at the exhaust port of the supercharger, with high exhaust density and small reactor volume, making the arrangement convenient.

3.3.3 NOx storage-reduction (NSR)
NOx storage reduction technology (NSR) refers to the adsorption of NOx by adsorbents (noble metals or alkali metals, etc.) and temporary storage in the form of nitrate or nitrite when the diesel engine is in the state of dilute combustion and the exhaust gas is in the state of oxygen enrichment. When a
diesel engine is oxygen-rich, the nitrates or nitrites stored in the adsorbent break down and release NOx. React with reducing agent CO, H2 and HC to produce N2.

NSR technology has the advantage of high reaction efficiency (60% ~ 80%), reaction conditions are relatively simple, small footprint, NO need to add reducing agent [32] in a row, but because of the diesel engine exhaust gas of SO2 and NO competition, and the reaction resistance to sulfur poisoning of the catalyst performance is poor, and the sulfur content in diesel oil is higher, so this technique only used in small diesel engines, applications in large Marine diesel engine has the certain difficulty[33].

3.3.4 Non-thermal plasma (NTP)
Non-thermal plasma technology refers to the use of low-temperature plasma to convert the gases in the exhaust gas into electrons, ions and free radicals, etc., and these particles form higher active oxides to promote the selective catalytic reduction reaction. Plasma denitration includes direct oxidation and indirect oxidation. Direct oxidation is the direct passage of waste gas through the discharge area, while indirect oxidation is the injection of a strong oxidizing substance (such as ozone) into flue gas. Indirect oxidation has a higher oxidation rate than direct oxidation [34,35].

Non-thermal plasma technology will produce pollution products NO2, HNO3 and so on, which can be combined with other technologies to not only deal with these pollution products, but also change the working mechanism to further reduce the NOx content in the tail gas. Non-thermal plasma technology with NOx storage technology +EGR technology can directly reduce NOx to N2, reducing the production of NOx [36]. Using dielectric barrier discharge technology to generate ions and combine with photocatalyst TiO2 to remove NO can effectively improve the removal efficiency of NOx [37]. MOK Y S et al. [38] found that the dielectric barrier discharge method was used to generate low-temperature plasma, which oxidized NO into NO2 and decomposed the microparticles. Then, under catalytic action, NO2 was reduced to N2, which could remove both PM and NOx.

Non-thermal plasma technology has the advantages of high catalytic efficiency, high reactant low temperature activity and insensitivity to sulfur [39], and can better meet the needs of production and application when combined with other technologies, but it is still in the research stage in the field of ship emission control.

3.3.5 Photocatalysis technology
Photocatalytic technology means that the photocatalyst oxidizes or reduces NOx and other harmful components in the exhaust gas into pollution-free substances under a certain amount of light. Generally, TiO2 is used as a photocatalyst and supported on ceramic, glass fiber, molecular sieve, silica gel or activated carbon, which can increase its surface area, improve the activity of TiO2 and increase the removal rate of NOx. The advantages of photocatalytic technology include low energy consumption, no additional reactants, no secondary pollution and mild reaction. Photocatalytic technology can also remove SO2 from diesel engine exhaust at the same time, but the inactivation of catalyst materials and low light utilization efficiency need to be solved urgently [40]. The combination of photocatalytic technology and Non-thermal plasma technology is better than the emission reduction effect of photocatalytic technology and Non-thermal plasma technology alone [41]. Fan guangli et al. [42] also found that under the conditions of flow rate of 1 L/min, O2 volume fraction of 4%, discharge voltage of 14 kV and uv lamp power of 15 W, the removal rate of NO reached 65%~75% when using photocatalytic technology and Non-thermal plasma technology, and the removal effect was better than that of Non-thermal plasma technology and photocatalytic technology alone.

4. Conclusion and prospect
With the development of Marine industry, NOx caused by Marine diesel engine is becoming more and more serious. IMO began to implement Tier III regulations in emission control areas as early as 2016. With the further deterioration of Marine ecological environment, more and more areas will be listed as emission control areas, Marine diesel engine NOx emission restrictions will be more stringent.
At present, NOx control technology of diesel engine mainly includes fuel pretreatment, dual-fuel engine, combustion chamber optimization, fuel injection system optimization, charge adjustment, humidification technology, SNCR, EGR, SCR, NSR, NTP, photocatalytic technology and so on. High temperature and oxygen-rich environment in the diesel engine cylinder will lead to a large amount of NOx, while combustion chamber optimization, fuel injection system optimization, charge regulation and other in-machine purification technologies are used to optimize the diesel engine combustion chamber structure, fuel injection system, intake and exhaust system, etc., with high cost and long development cycle. FWE, HAM and DWI have limited NOx removal effects and corrodes the diesel cylinder. SNCR technology requires no catalyst and has a low cost of control, but improper control can cause a large amount of ammonia to escape, leading to corrosion and blockage of the exhaust pipe. EGR has small structural changes, low cost and significant effect, but it will worsen combustion, increase fuel consumption and increase PM emissions. The catalyst in NSR technology has poor resistance to sulfur poisoning, so it is difficult to be used in large Marine diesel engines. NTP technology and photocatalytic technology have high catalytic efficiency, but they are still in the research stage in the field of ship emission control.

Dual-fuel engines and SCR technology can achieve more than 90% NOx removal, which can meet the Tier III standards in the IMO convention. Among them, the dual-fuel engine can not only reduce NOx emissions, but also reduce SO2 and CO2 emissions. Dual-fuel engines use natural gas as the main fuel, and can achieve a diesel substitution rate of more than 85%. In today's energy crisis and increasingly serious environmental pollution, it is of great significance to develop alternative energy and realize energy saving and emission reduction. However, in the practical application process, the reliability of dual-fuel engines needs to be improved.

SCR technology can reduce NOx emissions by more than 90% and can meet Tier III emission standards. For existing diesel engines that meet IMO Tier II emission limits, installing SCR post-processing equipment is the most commonly used means at present. However, the installation cost of SCR technology is high, the control strategy is complex, and the catalyst is easy to deactivate. Therefore, prolonging the service life of SCR catalyst and reducing the initial installation and operation cost of SCR catalyst are important directions for future development.

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