

DEVELOPMENT OF COAL FLUE GASES DENITRIFICATION TECHNOLOGIES IN CHINA

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Abstract. In the process of coal burning, a large amount of smoke will be produced, and there is a large amount of NO_x in the flue gas, only by removing these substances can the pollution degree of the flue gas be reduced. This paper analyzes the coal consumption and NO_x emission in China in recent years, and summarizes the industrial emission sources of NO_x. The principle, process flow, research status and development prospect of selective catalytic reduction (SCR), selective non-catalytic reduction (SNCR), ozone oxidation and absorption in traditional flue gas denitrification technology and complex absorption and photocatalytic oxidation in new flue gas denitrification technology are discussed in detail. The denitration rate, advantages and disadvantages of the traditional flue gas denitration technology in the denitration market are summarized. The technological characteristics and economy of the above five flue gas denitration technologies were compared, and the development direction of flue gas denitration technology in China was pointed out.

Keywords: coal-fired boilers, nitrogen oxides, emissions, denitration technology, catalyst.

1. Introduction

Environmental pollution and energy shortage are two major problems facing the world (He et al., 2021). Air pollution problems such as compound acid rain, photochemical smog and haze caused by NO_x

emission seriously endanger human health, destroy the ecological environmental balance, and restrict the rapid economic and social development in our country. In the past 30 years, coal consumption has accounted for about 70 % of China's primary energy consumption (National Bureau of Statistics, 2023). NO_x emissions from coal burning account for about 90 % of national emissions (Jianqiang et al., 2017).

Flue gas denitration technology is the most effective control method in the current flue gas denitration market (Ministry of Ecology and Environment of the People's Republic of China, 2023). Based on the background of ultra-low emission transformation in the coal power industry with the largest NO_x emission, this paper summarizes the traditional industrialized flue gas denitrification technologies in China's current flue gas denitrification market from the perspectives of NO_x emission reduction principle, application status and future prospects, and explores some new flue gas denitrification technologies in laboratory research stage. In order to provide reference for the future research and development of flue gas denitration technology.

Atmospheric pollutants NO_x mainly refer to NO and NO₂, among which about 95 % of NO_x emitted from coal burning is NO, and the rest is NO₂ (Jianhua, 2019). According to the annual report of

Environmental Statistics in 2022 (Ministry of Ecology and Environment of the People's Republic of China, 2023), China's NO_x emissions were 8.957 million tons, down 10.34 % from 2021, of which industrial sources, domestic sources and motor

vehicle emissions accounted for 37.21 %, 3.78 % and 58.80 %, respectively, and the rest were emissions from centralized pollution control facilities. The proportion of NO_x emissions from various industries in industrial sources is shown in Fig. 1.

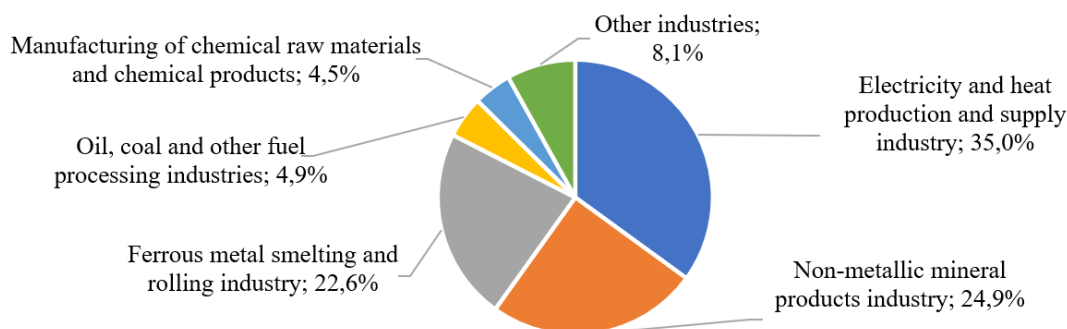


Fig. 1. The proportion of NO_x emissions in various industries in 2022

It can be seen from Fig. 1 that the three industries with large emissions in industrial sources are power, heat production and supply industry, non-metallic mineral products industry, ferrous metal smelting and rolling industry, accounting for 82.5 % of industrial source emissions. For the power industry with the largest NO_x emission, the Action Plan for Energy Conservation and Emission Reduction Upgrading and Transformation of Coal Power Generation (2024–2027) requires that by 2027, all coal power units with the conditions for transformation in China should achieve ultra-low emissions, that is, the NO_x emission concentration should not be higher than 50 mg/m³ (when the benchmark oxygen content is 6 %) (National Development and Reform Commission of the People's Republic of China, 2024). The implementation of the ultra-low emission policy in the coal power industry has accelerated the development of flue gas denitrification technology in China, and promoted the ultra-low emission transformation of cement, chemical and other industries to a certain extent.

2. Materials and Methods

In this study, we used literature analysis and other methods to study and explore the flue gas denitrification technology of coal-fired boilers. By reviewing the research status of traditional flue gas denitrification technology and new flue gas denitrification technology, the denitrification rate, principle, advantages and disadvantages and application status of SCR, SNCR, ozone oxidation absorption

denitrification method, complex absorption method and photocatalytic oxidation method were discussed, and the development direction of flue gas denitrification technology was prospected.

3. Traditional flue gas denitrification technology

The technology of flue gas denitration can be divided into dry method and wet method according to whether water is added in denitration process and the dry and wet state of the product. Traditional dry denitration methods mainly include selective catalytic reduction (SCR), selective non-catalytic reduction (SNCR), activated carbon, electron beam irradiation and pulse corona method. Traditional wet denitration mainly includes liquid phase absorption method and oxidation absorption method. In the following, SCR, SNCR and ozone oxidation absorption method, which are the three flue gas denitrification technologies with high application rate, are discussed in detail and some characteristics of the above technologies are compared and analyzed.

3.1. Selective Catalytic reduction Denitration Technology (SCR)

SCR technology is based on ammonia, liquid ammonia, urea and other reducing agents, supported by metal oxides as catalysts, NH₃ selectively catalyzed the reduction of NO_x to N₂ and H₂O(g) on the catalyst surface without O₂ oxidation in the flue gas. The SCR technology process flow is shown in Fig. 2.

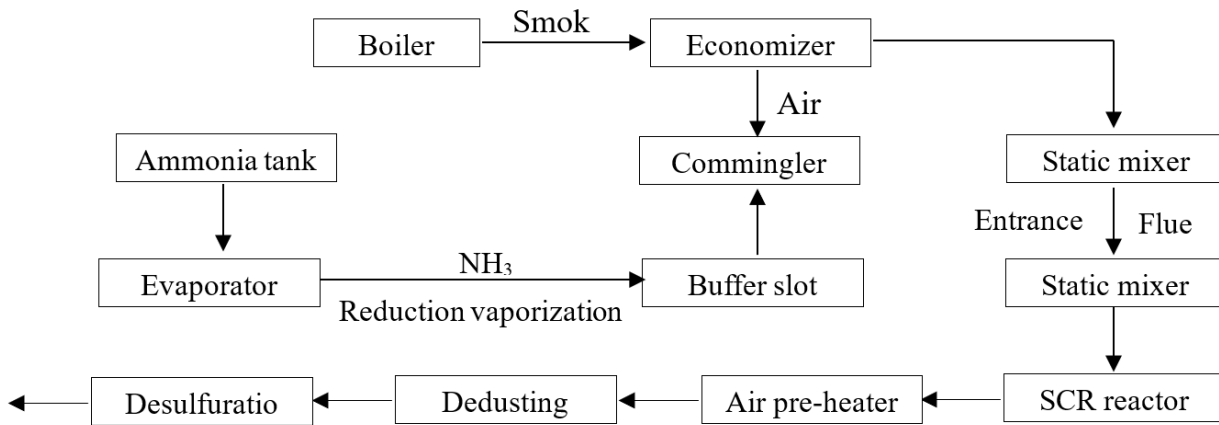
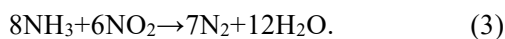
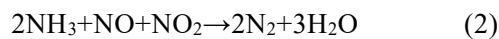
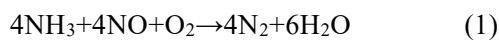


Fig. 2. The technological flow chart of SCR

In this process, the coal flue gas is discharged from the boiler into the economizer and mixed with NH_3 in the inlet flue of the SCR reactor. NH_3 comes from the liquid ammonia storage tank, and the liquid ammonia is evaporated into NH_3 by steam decompression into the buffer tank. Air is injected into the mixer to dilute pure ammonia and increase the oxygen content of the system. After NH_3 is fully mixed with air, it is sprayed into the reactor inlet flue through the jet grating. NH_3 and flue gas are evenly mixed by a static mixer and enter the main body of the reactor. At this time, NO_x is subjected to catalytic reduction under the action of catalysts and reducing agents, and the purified flue gas enters the air preheater for pre-heating, followed by dust removal and desulfurization. The reaction involved in this process is shown in formula (1)–(3).

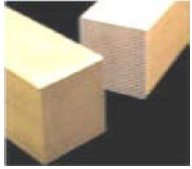




The main catalysts used in SCR denitration technology are V_2O_5 , Fe_2O_3 , CuO , Cr_2O_3 , etc., among which V_2O_5 has the highest catalytic activity. At present, the catalyst used is a vanadium titanium system dispersed on TiO_2 with V_2O_5 as the main active component and WO_3 or MoO_3 as the cocatalyst, namely $\text{V}_2\text{O}_5\text{--}\text{WO}_3/\text{TiO}_2$ or $\text{V}_2\text{O}_5\text{--}\text{MoO}_3/\text{TiO}_2$. The principle of catalytic reaction is that NH_3 is quickly adsorbed on the surface of V_2O_5 , reacts with NO , forms an intermediate product, decomposing into N_2 and H_2O , and the active point of the catalyst is

quickly recovered in the presence of O_2 , and continues the next cycle to carry out the chemisorption and reaction process. The reaction steps can be decomposed into: (1) NH_3 diffuses to the catalyst surface; (2) Chemical adsorption of NH_3 on V_2O_5 ; (3) NO diffuses to the catalyst surface; (4) NO reacts with NH_3 in adsorbed state to generate intermediate products; (5) The intermediate products decompose into the final products N_2 and H_2O ; (6) Outward diffusion away from the catalyst surface. According to the structure and external shape of the catalyst, there are three types: corrugated type, plate type and honeycomb type. The characteristics of the three catalysts are shown in Table 1. Because the reaction is sufficient, the honeycomb catalyst is the most used. SCR occupies an important position in flue gas denitrification technology because of its advantages of high denitrification efficiency and the most mature technology, but the problems such as high investment cost of vanadium-titanium catalysts and solid waste pollution can not be ignored. The waste vanadium and titanium catalysts can be recycled by washing with water, thermal reduction, acid solution and other methods (Wu et al., 2002), which can effectively slow down solid waste pollution and reduce investment costs, and achieve a win-win situation for both environment and economic benefits. The development of SCR catalysts in the future is still mainly based on the wide application of vanadium-titanium catalysts and deepening the research on their regeneration methods, and vigorously develop green and low-cost vanadium-free catalysts.

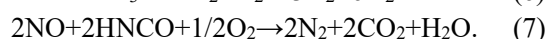
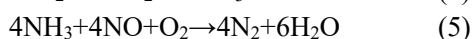
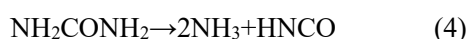
Table 1

Characteristics comparing three kinds of catalyst

Type	Base material	Picture	Technical characteristics
Cellular	Bulk extrusion		The overall medium is uniform, the specific surface area is large, the volume is small, the weight is light, the use is wide, the market share is high (about 70 %)
Plate-type	Stainless steel mesh		The specific surface area is small, the volume is large, the production cycle is short, and the ash is easy to accumulate between the upper and lower two catalysts
Ripple type	Fiber		Medium specific surface area, poor wear resistance, between the upper and lower two catalysts, easy to accumulate ash, low market share (not more than 5 %)

3.2. Selective Non-catalytic Reduction Denitration Technology (SNCR)

SNCR is a method of reducing NO_x to N_2 by injecting an amino reducing agent such as ammonia or urea into the boiler furnace or the circulating fluidized bed boiler separator at high temperature under the condition of no catalyst, and rapidly pyrolyzing or evaporating the reducing agent into NH_3 . The process is in the boiler outlet smoke temperature $850\sim 1100\text{ }^\circ\text{C}$, urea or ammonia and other reducing agents directly injected, in the case of high temperature, ammonia and the flue gas in the reaction of nitrogen oxides, nitrogen oxides reduction, reduce the concentration of nitrogen oxides. The main reactions involved in this technique are shown in (4)–(7).



SNCR technology has short construction period, simple construction, no need to replace the induced draft fan, less investment, insensitive to changes in coal, medium denitrification efficiency, and is suitable for the transformation of old boilers in China. The disadvantage of this technology is that the denitrification efficiency is low, and ammonia escape is easy to occur. Therefore, in terms of flue gas

denitrification, the use of SNCR technology alone is subject to some limitations.

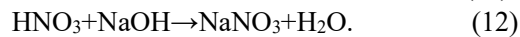
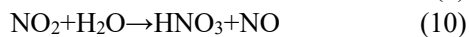
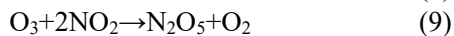
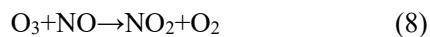
In the denitration process, no catalyst is used, and NH_3 escapes at $10\sim 15$ ppm. When spraying reducing agent into the furnace, because the reduction reaction of NO_x only occurs between $850\text{ }^\circ\text{C}$ and $1100\text{ }^\circ\text{C}$, the reaction temperature of reducing agent is one of the key factors for the reduction efficiency. The optimum reaction temperature is $950\text{ }^\circ\text{C}$. The key to the successful implementation of SNCR technology is that the reducing agent must be sprayed into the most effective temperature range in the furnace, and the total time of the reactants staying in the reactor should be controlled as much as possible, so that the reducing agent can be dispersed and mixed evenly with the flue gas at the most appropriate temperature, so as to maximize the utilization efficiency of the reducing agent and control the minimum ammonia escape.

SNCR process structure is relatively simple, no additional denitrification equipment is required, and is mostly used as a supplementary means of low nitrogen combustion or SCR. SNCR and SCR combined technology is a denitrification technology that integrates the advantages of high denitrification rate of SCR, low ammonia escape and low cost of SNCR. The escaped ammonia in SNCR can be further used as a reducing agent of SCR, effectively saving the consumption of reducing agent and catalyst, and thus reducing the operating cost.

3.3. Ozone oxidation absorption denitrification method

Ozone oxidation absorption and denitrification is a method of oxidizing the water-soluble NO in the flue gas into NO₂, N₂O₅ and other water-soluble substances, and then spray absorption with water, acid or alkali solution.

After dust removal, the flue gas oxidizes NO into high-priced NO_x with O₃ in the NO_x generator. The flue gas containing high-priced NO_x enters the absorption tower and contacts with NaOH solution to generate NaNO₃. After the reaction, the flue gas is purified by the demister and discharged into the atmosphere. Part of the reaction mixture is driven into the tower by the circulating pump for recycling, and part of the reaction mixture is discharged by the pump after filtration, separation and drying to obtain NaNO₃ crystals. The principle involved in this process is shown in equations (8)–(12).



The main factors affecting oxidation are the ratio of molar number between O₃ and NO and the oxidation time. Practice shows that the oxidation rate of NO increases with the increase of O₃/NO. Ozone denitrification rate is relatively high, the highest can reach more than 85 %. The main factors affecting ozone denitrification are: ozone amount, reaction time, mixing adequacy of ozone and flue gas, etc. Practice shows that the oxidation residence time of ozone in flue gas is a more important factor.

At present, domestic and foreign research on this method focuses on the study of oxidation and denitrification of O₃ based on wet flue gas desulfurization technologies such as calcium, magnesium and sodium. Sun et al. (Sun et al., 2015) used the combination of O₃ and MgO for denitrification, and the experiment showed that when the pH of solution was 6.5 and the concentration of MgO was 0.02 mol/L, the optimal removal rate of NO₂ was 75 %. Shaopeng et al., 2015 conducted desulfurization and denitrification experiments with O₃ and ammonia solution. The research shows that when pH is 10, the molar ratio of O₃/NO is 1, and the ammonia concentration is 0.3 %, the removal rates of SO₂ and NO_x can reach 99 % and 90 % respectively. Chunhu et al., 2014 used O₃ oxidation combined with Na₂S₂O₃ – NaOH solution wet spray to achieve NO removal. When the molar ratio of O₃/NO is 1.1~1.2, the concentration of Na₂S₂O₃ is 2 %, and the pH is 9, the denitrification rate can reach 75 %. Ozone oxidation absorption method has no secondary pollution, the process is simple, only need to install the ozone generator and other devices before the existing desulfurization equipment can achieve simultaneous desulfurization and denitrification, especially suitable for SCR and SNCR can not carry out low-temperature denitrification field (< 200 °C); However, the preparation cost of the required ozone is high, and the future should focus on the study of energy-saving and efficient ozone generators to reduce the investment and operating costs of the system.

3.4. Comparison of characteristics of traditional flue gas denitration technology

Table 2 shows the comparison of characteristics of traditional flue gas denitrification technologies.

Table 2

Comparison of characteristics of traditional flue gas denitrification technology

Technology	Denitration rate	Principle	Merits and demerits	Application status
1	2	3	4	5
SCR	80~90 %, up to 95 %	NO _x was reduced to non-toxic N ₂ and H ₂ O by amino reducing agent under catalyst	High desulfurization rate, the most mature technology; However, the investment and operation costs are high, the catalyst is expensive and prone to poisoning, and accompanied by ammonia escape	Widely used; It is mostly used in the coal-fired power industry with large emissions

Continuation of Table 2

1	2	3	4	5
SNCR	> 60 %, up to about 75 %	NO _x is directly reduced to N ₂ and H ₂ O by amino reducing agents	Mature technology, low investment and operating costs; Ammonia escape is serious	Widely used; It is mostly used in the coal-fired power industry with large emissions
Oxidation absorption method	> 85 %, up to 90 % (O ₃ method)	NO is oxidized to NO ₂ by gaseous phase (O ₃ , Cl ₂ , etc.) or liquid phase (NaClO ₂ , HClO ₃ , H ₂ O ₂ , etc.) oxidants, and then removed by re-absorption	O ₃ method is simple to operate, but the investment and operation cost are high; NaClO ₂ , HClO ₃ expensive, easy to corrode equipment; H ₂ O ₂ is easy to decompose and consumes a lot of light	Only O ₃ oxidation method is widely used, mostly used in petrochemical, carbon and other industries; The rest are not widely used in industry
Adsorption method	80~90 % (Activated carbon method)	The adsorbent (activated carbon/coke, molecular sieve, etc.) catalyzes and adsorbs NO _x and then reduces it with amino, and the adsorbent is regenerated by heating or steam	The equipment is simple, can cooperate desulfurization; But the adsorption amount is small, the amount of adsorbent is large, and the regeneration is frequent	Activated carbon/coke method is widely used. Suitable for industries with small emissions
Plasma method (electron beam, pulse corona)	> 70 %	The free radical produced by high-energy electrons is used to oxidize NO to NO ₂ , and then into NH ₃ and H ₂ O (g) to produce ammonium nitrate	Simple operation, can cooperate with desulfurization and dust removal, no waste waste water; But high energy consumption	Not widely used
Liquid phase absorption method	> 90 %	Use common water, lye (caustic soda, lime milk, ammonia, etc.) or acid (nitric acid, sulfuric acid, etc.) to absorb and remove NO _x	The by-product with high concentration of nitric acid / (nitrite) nitrate can be recycled. However, a high NO ₂ /NO is required	Widely used; It is mostly used in high-concentration NO ₂ exhaust industries, such as nitric acid plants

4. New flue gas denitrification technology

According to the wet method, the new flue gas denitrification technology can be divided into complex absorption method, liquid film method, microbial method, etc. Dry methods are mainly photocatalytic oxidation method, electroassisted catalysis method, etc., such technologies are mostly in the experimental research stage, and the following will be

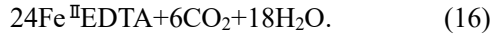
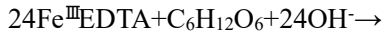
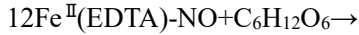
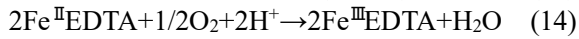
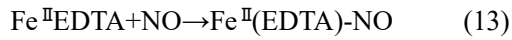
specifically discussed for complex absorption method and photocatalytic oxidation method.

4.1. Complex absorption method

The complexation absorption method is to use the coordination between the liquid phase complexation agent and NO to absorb NO, and then use microorganisms, electrolysis or adding reducing

agents to reduce the complexation agent that absorbed NO, so as to regenerate and recycle the method. At present, the most widely studied process at home and abroad is the removal of NO by the Fe^{II}EDTA chelating agent composed of the ligand ethylenediamine tetraacetic acid (EDTA) combined with biological reduction method (He et al., 2018).

In the process of denitrification, Fe^{II}EDTA and NO are complexed to form ferrous nitrosyl complex, which makes NO enter the liquid phase. Secondly, the denitrifying bacteria reduced Fe^{II}EDTA-NO to N₂ under the organic carbon source such as glucose and then multiplied themselves, while Fe^{II}EDTA was regenerated. In addition, there is a side reaction of Fe²⁺ being oxidized to Fe³⁺ by O₂ in the flue gas. The process reaction is shown in equation (13)–(16).

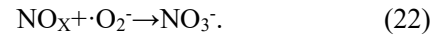
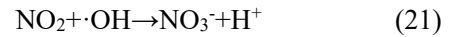
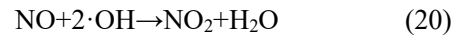
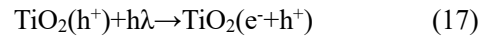


In view of the problems of complex agent consumption and low denitrification rate caused by NO complexation absorption capacity of Fe³⁺ generated in the denitrification process, relevant scholars have conducted a lot of research on how to effectively prevent Fe²⁺ oxidation and efficiently regenerate absorptive liquid. Wang Fei et al. (Wang, 2015) carried out nitrogen removal experiments of Fe^{II}EDTA in a rotating packed bed by using super-gravity technology. The study showed that when the super-gravity factor β was 82, the decrease of NO removal rate was only 12.9 %, much lower than 74.2 % ($\beta = 10$), which verified that the rotating packed bed could effectively avoid Fe²⁺ oxidation. Zhang Yu et al. (Yu et al., 2017) studied the simultaneous removal of Fe^{II}EDTA-NO and Fe^{III}EDTA by denitrifying bacteria in an anaerobic reactor with sodium lactic acid carbon source. The results showed that when the hydraulic retention time was 16h and pH was 7, the average removal rate of Fe^{II}EDTA-NO could reach 96.61 %. Jun et al. (Jun et al., 2018) proposed a nitrogen removal reaction of Fe^{II}EDTA in a biological rotary drum filter and studied the NO mass transfer model. Long-term operation results showed that the denitrification rate of the reactor could be stabilized at about 95 %, and the average relative deviation between the model and the experimental data was only 2.27 %. The complex absorption combined with biological

reduction method has the advantages of fast reaction rate, recyclable absorption solution, no secondary pollution and simultaneous desulfurization and denitrification. However, due to the oxidation of complex agents and the anaerobic nature of microorganisms, the industrial application of this method is hindered. In the future, the method should focus on cultivating high-efficient aerobic microorganisms and optimizing microbial regeneration equipment to improve the denitrification rate and regeneration rate of the absorption solution.

4.2. Photocatalytic oxidation method

Photocatalytic oxidation is a method of denitrification in which the photocatalyst is stimulated by energy to produce electron-hole pairs under certain light conditions, and the electrons and holes form superoxide and hydroxyl radicals with O₂ and H₂O(g) in the flue gas respectively, and then the NO_x adsorbed on the surface of the catalyst is oxidized to nitric acid. The reaction mechanism involved in this method is shown in equations (17)–(22).



At present, the commonly used photocatalysts at home and abroad are TiO₂, ZnO and CdS semiconductors, among which nano TiO₂ has been widely studied for its advantages of green environmental protection, high catalytic activity, and good chemical and thermal stability. However, it also has the disadvantages of only absorbing ultraviolet rays in sunlight ($\lambda < 387 \text{ nm}$) and low utilization of visible light ($380 \text{ nm} < \lambda < 780 \text{ nm}$), and easy rapid recombination of electron-hole pairs. In this regard, relevant scholars proposed that the visible light absorption spectrum can be broadened by means of precious metal modification, metal or non-metal doping, semiconductor composite, etc., and the hole-electron pair recombination can be suppressed to improve the nitrogen removal rate (Ye et al., 2016, Xiaoming et al., 2018). Yang Wei (Chunhu et al., 2014) prepared rGO – TiO₂ / ASC catalyst using active semi-coke (ASC) as the carrier, and the results showed that the addition of rGO could effectively inhibit the electron-hole recombination, and the

denitrification performance of the photocatalyst remained basically unchanged after three times of water vapor regeneration. Hu Jing (Jing et al., 2017) obtained blackTiO₂ with the function of widening absorption spectrum through TiO₂ self-doping and prepared Bi / BiOI / blackTiO₂. The denitrification rate of the catalyst could reach 70 % under simulated illumination, and the activity of the catalyst could still reach 68 % in 5 regeneration experiments.

Photocatalytic oxidation for denitrification has the advantages of mild reaction conditions, co-desulfurization and no secondary pollution (Shuai et al., 2018), but the problems of high cost and low efficiency caused

by the narrow absorption spectrum of visible light caused by the photocatalyst limit the industrial application of this method. In the future, it is still necessary to broaden the research on visible light absorption spectrum and develop new photocatalysts.

Table 3 compares and summarizes the characteristics and economy of the above three traditional flue gas denitrification technologies, SCR, SNCR and O₃ oxidation absorption method, and two new flue gas denitrification technologies, complex absorption method and photocatalytic oxidation method. The specific contents are shown in Table 3.

Table 3

Comparison of characteristics of traditional flue gas denitrification technology

Index	SCR	SNCR	O ₃ oxidation absorption method (1 million m ³ /h smoke volume)	Complex absorption method	Photocatalytic oxidation
Sort	Dry method	Dry method	Wet process	Wet process	Dry method
Investment cost /(RMB yuan ·kW ⁻¹)	100~150	30~40	50 million yuan	—	—
Running cost /(RMB fen·kWh ⁻¹)	0.7~1.2	<0.0035	16 yuan/kg(NO _x)	—	—
Secondary pollution	NH ₃ 、Waste catalyst	NH ₃	none	none	none
Ammonia escape rate /(mg·L ⁻¹)	3~5	10~15	none	none	none
Simultaneous desulfurization	no	no	yes	yes	yes
Denitration product	N ₂	N ₂	NO ₃ ⁻	N ₂	NO ₃

5. Conclusion

The transformation and upgrading of traditional flue gas denitrification technologies such as SCR and SNCR have made coal-fired power, cement and chemical industries achieve ultra-low emissions. Although such technologies are mature in industrial application and have high removal efficiency, there are serious problems such as nitrogen resource waste and gas-solid secondary pollution, especially waste catalysts in SCR. The complex absorption method developed to solve these problems has the characteristics of no secondary pollution and can cooperate with desulfurization. The photocatalytic oxidation method can recover and apply the nitrogen resources in flue gas, which meets the dual needs of environmental and economic benefits.

In the future, the existing efficient SCR, SNCR and ozone oxidation absorption technologies will be widely used in flue gas denitrification technology, and the integration of desulfurization and denitrification technologies such as adsorption method and plasma method will be vigorously developed. New flue gas denitrification technologies such as complex absorption method and photocatalytic oxidation method will be deeply studied, and flue gas denitrification technologies suitable for local characteristics will be explored.

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