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Research progress of flue gas desulfurization and denitrification technology in coal-fired power plants

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Abstract

As NO_X and SO_2 emissions during coal-fired power generation lead to air pollution such as acid rain and haze, NO_X and SO_2 emissions must be controlled according to the national requirements. In this paper, the research status of desulphurization and denitrification technology is summarized, the classification, principle, advantages and disadvantages of flue gas desulphurization technology, flue gas denitrification technology and integrated flue gas desulphurization and denitrification technology are reviewed respectively, and the development direction of flue gas desulphurization and denitrification technology is prospected.

Keywords

Coal-fired flue gas; desulphurization; denitrification; integration.

1. Introduction

China is a large country with a large population, and it has a great demand for electricity. Therefore, many power generation methods have emerged, such as thermal power generation, hydropower generation, wind power generation, nuclear power generation, etc. Among them, thermal power generation accounts for 72% of Chinese power generation method, which is the main power generation method in China at the present stage. Thermal power generation is mainly powered by coal combustion. However, coal combustion releases large amounts of sulfur dioxide (SO_2), nitrogen oxide (NO_X) and dust into the atmosphere. Most of the NO_X are NO and NO_2. The emission of these harmful gases and dust leads to the continuous appearance of air pollution such as acid rain and haze, which seriously affects people's normal life. The air pollutant emission standards for thermal power plants that have been implemented since January 1, 2012 have further increased the requirements for the reduction of SO_2 and NO_X. The emission limits of sulfur dioxide and nitrogen oxides in the "Emission Standard of Air Pollutants for Thermal Power Plants" (GB13223-2011) are shown in Table 1. From the perspective of Chinese energy demand, coal-fired power generation will still occupy the main position of electricity production for a period of time in the future, so the control of pollutant emissions from coal-fired power plants is the top priority of air pollution.

Table 1 Limits of air pollutants emission from thermal power boilers

Types of fuel and heat conversion equipment	Contaminant project	Application condition	Limiting value(mg/m^3)	Key area limit(mg/m^3)	Pollutant discharge monitoring location
Coal-fired boiler	SO_2	New boiler	100-200	50	Chimney / flue
		Existing boiler	200-400		
	NO_X	All	100-200	100	

2. Research and development status of flue gas desulfurization and denitrification

2.1. Development status of flue gas desulfurization

Sulfur dioxide gas is one of the main factors leading to the formation of acid rain. SO_2 reacts with water vapor in the air to form H_2SO_3, which is oxidized to H_2SO_4 by O_2 in the air. Acid rain can lead to soil acidification, poisoning and even death of plants. Damage to buildings, cultural relics and industrial facilities. Dissolved in drinking water, causing harm to people's health. Faced with the serious impact of sulfur dioxide pollution, and increasingly stringent national emission requirements. More than 200 flue gas desulfurization technologies have been developed in various countries in the world, but only a dozen can be applied to the actual desulfurization process [1].

Desulphurization technology can be divided into desulphurization before combustion, desulphurization during combustion and desulphurization after combustion, among which desulphurization after combustion is the most widely used method of desulphurization in the world, also known as flue gas desulphurization (FGD). According to the wet and dry state of absorbent and desulfurization products in desulfurization process, desulfurization technology can be divided into wet method, dry method and semi-dry method. Among them, wet FGD technology is desulphurization and treatment of desulphurization products with absorbent solution or slurry in wet state, which is the most widely used desulphurization method at present. Limestone-gypsum method in wet process is the most mature and most widely used desulphurization method among many desulphurization methods, with a desulphurization efficiency of up to 90%. As early as in Japan, the United States and Germany desulfurization applications reached 75 %, 80 % and 90 %, respectively, while in China the proportion of desulfurization applications also reached 90 %. The most widely used commercial technique in the world is the calcium method, which accounts for more than 90%. Although wet desulfurization is highly efficient and stable in the market, long-term operation of the system is prone to corrosion, scaling, waste water and residue generation, environmental pollution, low recovery rate of by-products and other problems make wet FGD lose its advantages in environmental protection and economic issues [2].

At present, the main flue gas desulfurization technologies are divided into the following five types: calcium method based on $CaCO_3$ (limestone), magnesium method based on MgO, double alkali method based on NaOH, ammonia method based on NH3 and circulating fluidized bed flue gas desulfurization [3].

Also known as calcium desulphurization limestone - gypsum flue gas desulfurization, lime writing broken ground into powder mixed with water and stir into the absorption slurry, in the absorption tower, absorption slurry mixed with flue gas contact, sulfur dioxide in flue gas with the calcium carbonate in the slurry and the oxidation of blowing air for chemical reaction to be removal, the final reaction products for plaster [4]. The removal efficiency of this method is as high as 90%, the absorbent is common, but the cost of primary input is high, and it is easy to scale, corrode, and cause serious secondary pollution by waste water and slag.

In the magnesium desulfurization method, MgO reacts with water to produce $Mg(OH)_2$, and $Mg(OH)_2$ neutralizes with SO_2 to produce $MgSO_3$, which is then oxidized by O_2 to $MgSO_4$. In terms of chemical reactivity, MgO is much larger than calcium-based desulfurizer, and its molecular weight is smaller than $CaCO_3$ and CaO. Therefore, under the same other conditions, the desulphurization efficiency of magnesium oxide is higher than that of calcium method. In general, the desulphurization efficiency of MgO can reach more than 95-98%, and the by-products can be recovered and reused. However, there are problems of waste water treatment and high temperature flue gas that are easily corroded and desulfurized [5].

The double alkali desulfurization method USES NaOH solution as the starting desulfurizer. The prepared NaOH solution is directly driven into the desulfurization tower to wash and remove SO_2 in the flue gas, and then the desulfurization product is replaced with limestone solution $Ca(OH)_2$ to generate NaOH and then returned to the desulfurization tower for recycling. he desulfurization efficiency of this method is above 90%, and the corrosion and blockage of pipelines and equipment in the circulating process of the system are relatively light. However, Na_2SO_4 is difficult to regenerate, so the amount of NaOH needs to be increased [6]. Therefore, this method is only suitable for flue gas desulfurization of sulfur coal boilers in small and medium-sized power plants.

Ammonia desulphurization is the reaction of ammonia or ammonia water with acidic SO_2 to produce $(NH4)_2SO_4$. In the absorption tower, the absorption solution is mixed with the flue gas in contact, and SO_2 in the flue gas reacts with (NH4)2S03 and NH_4HSO_3 in the solution to be removed, and the final reaction product is ammonia sulfate crystal. The desulphurization efficiency of this method is up to 95%, no waste residue wastewater, high heat utilization rate, can realize simultaneous desulphurization and denitrification, and its desulphurization by-product ammonium sulfate is a kind of agricultural fertilizer in some specific areas. But the price of ammonia is high, and volatile, resulting in corrosion problems.

The circulating fluidized bed method USES calcareous (CaO) as desulfurizer, CaO reacts with water to form $Ca(OH)_2$, $Ca(OH)_2$ reacts with SO_2 to achieve flue gas desulfurization effect. After the flue gas is cooled at high temperature, it is accelerated into the tower through the venturi device at the bottom of the fluidized bed tower, and reacts with the lime slurry to produce calcium sulfites and a small amount of sulfites. The solid particles after the reaction are removed from the bed, and the desulfurized ash returns to the tower to continue to participate in the reaction [7]. The turbulent state of the fluidized bed provides continuous particle contact for the circulation ratio of the system. The collision between the particles causes the reaction products on the surface of the absorbers to wear and flake constantly, which greatly improves the utilization rate and desulfurization efficiency of the absorbers. This method has the advantages of high desulfurization efficiency, no waste water and low operating cost, and has been widely used in the flue gas desulfurization of thermal power plants. However, the desulfurization process needs to use high purity and active lime as desulfurizer, and the comprehensive utilization of desulfurization products is also limited to some extent.

2.2. Development status of flue gas denitrification

The survey shows that 56% of NO_X emissions come from the combustion of coal, and the vast majority of coal is used for power generation in power plants. The nitrogen oxides emitted by coal-fired power plants are mainly nitrogen monoxide (NO) and nitrogen dioxide (NO_2), of which NO accounts for 95%, NO_2 accounts for 5%, and a small amount of nitrous oxide (N_2O)[8]. The harm of nitrogen oxide lies in that it can produce a series of photochemical reactions with hydrocarbons (HC) under solar irradiation: $NO+HC+O_2+$ sunlight $\rightarrow NO_2+O_3$, generating photochemical smog (NO_2+O_2). Photochemical smoke can cause red eyes, cough, sore throat, moist skin and other symptoms, and in severe cases, heart and lung failure. Nitrogen oxides combine with water in the air to eventually convert to nitric acid and nitrates, which are one of the causes of acid rain and cause damage to plants, crops and buildings. N_2O in nitrogen oxide reacts with O to form NO, NO reacts with ozone (O_3) to form NO_2, and NO_2 reacts with O to form NO. In this way, O_3 is repeatedly decomposed and destroyed the ozone layer. In order to control NO_X emissions, the state has also formulated relevant policies. In 2011, the 12th Five-Year Plan included NO_X in the binding index system for the first time before a large number of denitrification equipment began to be built. Therefore, Chinese denitrification industry started relatively late, and by the end of 2017, it had been put into use for flue gas denitrification of 98.4%. In 2012, the air pollutant emission standards for coal-fired power plants were implemented, and the 13th Five-Year Plan for 2016 proposed a 15% reduction in nitrogen oxide emissions compared with 2015.

There are two main way to reduce nitrogen oxide emissions, one is before burning treatment, namely the low nitrogen combustion technology, mainly by improving the combustion condition and improve combustion equipment, such as reducing excess air, combustion, flue gas recirculation combustion and low temperature air preheating method, though simple economic effective, but low NO_X removal efficiency, generally about 50% [9]. The other is the flue gas denitrification after combustion tail gas treatment, which is more efficient and widely used. Flue gas denitrification after combustion can be divided into dry flue gas denitrification and wet flue gas denitrification. Dry flue gas denitrification technology includes selective catalytic reduction (SCR) flue gas denitrification technology, selective non-catalytic reduction (SNCR) flue gas denitrification technology and SCR-SNCR mixed flue gas denitrification technology. The wet flue gas denitrification technology includes oxidation absorption method, alkali liquor absorption method and reduction absorption method [10].

Selective catalytic reduction (SCR) flue gas denitrification technology USES NH_3 as a reducing agent to reduce NO_X into N_2 and H_2O under the action of catalyst, which can accelerate the reaction speed and the temperature is between 315 ℃ and 400℃. Selectivity means that under the action of catalyst and the presence of oxygen, NH_3 preferentially reduces reaction with NOx, rather than oxidizing reaction with oxygen in flue gas. The denitrification efficiency of this method is 80%~90%, the technology is mature and widely used, the secondary pollution is small, but the investment cost is high, the operation cost is high, especially the consumption cost of reducing agent (ammonia water, liquid ammonia, urea, etc.) is high.

Selective non-catalytic reduction (SNCR) denitrification of flue gas is a method for NOx removal in the range of 850℃~1100℃ without catalyst. NO_X is reduced to N_2 and H_2O by reducing agent ammonia or urea. This method controls the temperature strictly. When the temperature is higher than the reaction temperature, NH_3 will be oxidized to NO. When the temperature is lower than the reaction temperature, the reaction is incomplete and the escape rate of ammonia is relatively high, leading to low efficiency [11]. Compared with SCR flue gas denitrification technology, SNCR flue gas denitrification technology system is simple, less investment, but its efficiency is 40%~50% lower than SCR technology, and its practical application is relatively less.

Mixed SNCR and SCR flue gas denitration technology is a combination of selective catalytic reduction (SCR) high denitration efficiency and selective non-catalytic reduction (SNCR) economic low cost advantage, through the NOx and reagent (NH_3) or urea reaction realize preliminary denitration, reduce NOx in flue flow content, escape again with ammonia removal of NOx in catalytic reduction reaction to produce harmless N_2 and H_2O [12]. Mixed SNCR-SCR flue gas denitrification technology not only maintains the high efficiency of denitrification, but also reduces the initial investment and operating costs, reduces the size of the catalyst and the pressure drop on the catalyst, and under the action of the catalyst, the escaped ammonia and NOx rereact to reduce the amount of ammonia escape. However, in the actual production process, mixed SNCR-SCR flue gas denitrification technology has certain deficiencies, the amount of escaped ammonia is difficult to control, and it is difficult to give consideration to the ideal high ammonia escape rate while maintaining the high denitrification efficiency. The control of the amount of escaped ammonia will affect the whole denitrification process and denitrification efficiency. Mixed SNCR-SCR technology requires SNCR to produce a large amount of escaped ammonia, which needs more reducing agent to consume.

The flue gas denitrification technology of oxidation absorption method is a flue gas denitrification technology that converts water-insoluble NO oxidation into water-soluble NO_2 and other high-priced NOx and USES alkali liquor absorption [13]. The oxidants commonly used

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in the oxidation absorption method are O_3 and H_2O_2, and the oxidation products of both are H_2O and O_2, which will not cause unnecessary pollution to the absorption solution [14,15]. H_2O_2 is chemically active and easy to decompose, limiting its large-scale use [16]. O_3 oxidation can oxidize NO in flue gas at room temperature and pressure without heating or pressurization, etc. Therefore, when O_3 content is sufficient to oxidize flue gas, high denitrification efficiency (≥90%) can be obtained. Therefore, this technical method has the advantages of low requirement for reaction temperature and high denitrification efficiency [17]. However, ozone generator is not suitable for small and medium-sized enterprises due to its large investment and high operating cost.

The basic principle of flue gas denitrification technology with alkali absorption method is to dissolve NOx and other nitrogen oxides in water to produce acidic solution, and then use alkali solution for neutralization reaction to treat nitrogen oxides in flue gas into nitrate and nitrite products with certain economic benefits [18]. The most commonly used lye absorption methods are $Ca(OH)_2$ and $CaCO_3$, both of which are economically and easily available. This method has low economic operation cost and is widely used in wet flue gas desulfurization and denitrification technology. Lye absorption also has certain defects. If NO catalyst is used, NO with the highest content in the flue gas is not easily soluble in water and alkaline solution, and it is difficult to remove NO in the flue gas [19,20].

Reduction absorption method flue gas denitrification technology is the use of activated carbon and other catalysts and the oxygen contained in the waste gas as an oxidant, NOx in the flue gas catalytic oxidation, and then through the absorption of liquid reducing agent N_2 reduction. The advantages of this method are low economic operation cost, simple process equipment and cheap catalyst. However, its disadvantages are also obvious. This method is difficult to remove NOx in flue gas, resulting in a low NOx removal rate and a slow reduction rate of NO by reducing agent. Therefore, it is necessary to improve the oxidation degree of NO and oxidize it to high-priced NOx [21,22].

2.3. Development status of flue gas desulfurization and denitrification integration

To sum up, at this stage, desulfurization and denitrification are all treated separately. Although it can effectively desulfurize and denitrify, due to the huge equipment, complex technology, and high cost, it affects the development of thermal power plants [23]. Therefore, on the basis of the traditional process, relevant technicians have developed new desulfurization and denitrification equipment and technology, which can simultaneously remove sulfur dioxide and nitrogen oxide in flue gas, and can not only treat flue gas, but also promote the development of thermal power plant. Integration of flue gas desulfurization denitration technology is divided into combined desulfurization and denitration technology desulfurization denitration technology at the same time, combined desulfurization denitration technology refers to the two cell removal of SO_2 and NO_X, using the most sophisticated of flue gas desulfurization technology at home and abroad (FGD) and selective catalytic reduction (SCR) technology with the combination of methods, but there are high investment, equipment covers an area of large, complex management and so on. At the same time, desulfurization and denitrification technology refers to the technology of NO_X and SO_2 removal in the same unit, which makes up for the shortcomings of combined desulfurization and denitrification technology and has become the focus of domestic and foreign research. Although this technology is still in the research stage, it has a great application prospect. Desulfurization and denitrification technology can also be divided into dry simultaneous desulfurization and denitrification technology and wet simultaneous desulfurization and denitrification technology.

The dry desulfurization and denitrification technologies mainly include activated carbon adsorption, electron beam, pulse corona and CuO adsorption. Simultaneous desulfurization and denitrification by wet method mainly includes complexation absorption, oxidation absorption and urea reduction absorption [24].

The removal of pollutants by activated carbon adsorption method is based on the large surface area of activated carbon, good porous structure, rich surface groups, as well as load performance and reduction performance, so it can be used as a carrier and reductant to participate in the reaction. There are mainly two processes, (a) SO_2 and NO_X are adsorbed by activated carbon, and chemical reactions take place between SO_2, H_2O, NO_X and O_2 in the flue gas. After being oxidized by O_2, SO_2 reacts with H_2O to form H_2SO_4. (b) Then an appropriate amount of ammonia is added in the reaction, and NO_X is directly oxidized and reduced to N_2 and H_2O. The removal efficiency of SO_2 and NO_X in this reaction is about 90% and 80% respectively. The reaction temperature is low, materials are easy to be obtained, and there is no secondary pollution. However, there are problems such as difficult regeneration of adsorbent, high cost of regeneration, high cost of adsorbent, and immature technology.

Electron beam method (EBA) and pulsed corona method (PCDP) belong to plasma method. Electron beam method is a kind of desulfurization and denitrification technology combining physical and chemical technology. Its basic principle is to use high-energy electron beam to irradiate flue gas and make it produce a variety of active groups to oxidize SO_2 and NOx in flue gas to produce HNO_3 and H_2SO_4, and finally react with NH3 added to flue gas to produce NH_4NO_3 and $(NH_4)2SO_4$[25]. The SO_2 removal efficiency of this method is above 95%, and the NOx removal efficiency is 80%~85%. Moreover, the equipment is simple and the by-products can be used as fertilizer. However, this method is not suitable for popularization because of its high power consumption, high operating cost and harmful radiation to human body. The pulsed corona method has the same desulfurization and denitrification principle in the electron beam method, but the difference is that the high-energy electrons are generated in different ways. The electron beam method is irradiated by the electron beam, while the pulsed corona method is radiated by the high-voltage power source. The extraction efficiencies of SO_2 and NO_X by pulsed corona method are above 90% and 50% respectively, which are in the small-scale experimental study stage [26], with low denitrification efficiency and also high energy consumption.

The study of CuO adsorption method [27] began in the 1980s, and the process of simultaneous desulfurization and denitrification is divided into two steps. The first step is to react CuO with SO_2 and O_2 in flue gas to generate $CuSO_4$ under the temperature of 300℃~450℃. The second step is to take $CuSO_4$ as catalyst and add NH_3 into the reaction to react with NO_X and O_2 to produce N_2 and H_2O. The efficiency of desulfurization and denitrification is 90%~95%, and the by-products can be recovered to reduce the secondary pollution. Although the adsorbent can be regenerated, the activity of the regenerated adsorbent decreases, and the desulfurization and denitrification efficiency in the later stage decreases.

Complexation absorption method is by adding metal complex reaction with NOx to produce metal nitrite compound to increase the solubility of NOx in the solution, and then use reducing agent to reduce NOx into ammonia and nitrogen, combined with wet flue gas desulfurization technology, to achieve the purpose of removing NOx and SO_2 at the same time [28,29]. At present, the most studied chelating agents are ferrous chelating agents and cobalt chelating agents, whose removal efficiency of NOx and SO_2 is as high as 85% and 95% under the best experimental conditions [30]. This method has high desulfurization and denitrification efficiency and no secondary pollution. However, there is certain instability in the operation process, and absorption liquid discharge is difficult to achieve efficient recycling, without good economy, so it will not be popularized in a short time.

Oxidation absorption desulfurization and denitrification is carried out on the basis of wet desulfurization and denitrification. Generally, oxidizing agent is used to oxidize NOx and SO_2 in the gas into NO_2 and SO_3, and then alkaline absorbent is used to remove them. Commonly used oxidants are $NaClO_2$, H_2O_2, O_3, $KMnO_4$, etc. he most studied method is the simultaneous oxidation of $NaClO_2$/NaClO [31,32], whose reaction process is relatively complex. To sum up, SO_2 generates HSO_3^- through hydration reaction in dissolved water, and ClO_2^-/ClO^- in the composite solution oxidized HSO_3^- to SO_4^{2-} and Cl-. ClO_2^-/ClO^- reacts with NO to produce NO2- and ClO^-, which in turn promotes the oxidation of NO_2^- to NO_3^-. The desulfurization and denitrification efficiency of this method is as high as 90%, but the concentration of $NaClO_2$ and NaClO ($NaClO_2$ (6mmol/L), NaClO (1mmol/L) and the PH of the solution are 5.5 to achieve maximum desulfurization and denitrification. Therefore, the complexity of the reaction and the accuracy of the control increase the operating cost in the later stage.

Urea reduction absorption method [33] takes advantage of the strong reductibility of urea to remove NOx and SO_2. The basic principle of the reaction is as follows: SO_2 and urea undergo hydrolysis reaction in water to produce SO_3^{2-} and NH_4^+ reaction to produce (NH_4)2SO_3. $(NH_4)_2SO_3$ is oxidized by O_2 in flue gas to $(NH_4)_2SO_4$, which can be recycled. Urea and NOx undergo an oxidation-reduction reaction to generate N_2, H_2O and CO_2. This method has small secondary pollution, simple process, but low efficiency. The desulfurization and denitrification efficiency under optimal operating conditions are 98% and 48%, respectively [34].

3. Conclusion and prospect

With the continuous development and innovation of science in recent years, more and more advanced technologies have been developed and put into production. At the same time, it needs to be understood that most of the current technologies are still in the experimental stage, and factors such as cost have caused the integrated flue gas desulfurization and denitrification technology to not be fully put into production. In future research, if the cost and regeneration of the adsorbent in the integrated process can be solved, it will inevitably promote the transformation of flue gas desulfurization and denitrification technology in thermal power plants around the world, which will have an inestimable effect on the control of air pollution in China.

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