Progress of Desulfurization and Denitration Technology of Flue Gas in China

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Abstract. The global environmental situation is becoming more and more serious. Coal combustion is one of the main sources of air pollution. When coal is burned, it will often produce a large number of sulfides and nitrates, which will cause much pollution to the environment. China has the largest coal consumption and production volume in the world, of which more than 70% of coal is directly used for combustion, and the resulting environmental problems have attracted the attention of the whole society. Therefore, in order to protect the environment, the desulfurization and denitration of flue gas in the combustion process of coal has become an important step. This work summarizes the desulfurization and denitration technology of coal-fired power plants in China. Firstly, it summarizes the existing desulfurization and denitration technologies, then summarizes the research progress of the combined desulfurization and denitration technology in coal-fired flue gas, and forecasts the development of future desulfurization and denitration technologies.

1. Energy utilization in China  
With the rapid development of China economy, energy consumption has also risen rapidly. According to the latest China Statistical Yearbook published by the National Bureau of Statistics in 2018, China's energy has consumed 4.49 billion tons of standard coal in 2017, 7.9 times that of 1978[1]. However, the energy reserves in China are unevenly distributed extremely, coal resources are relatively abundant, proven reserves rank third in the world, but oil and natural gas reserves are relatively scarce, and per capita reserves are less than 8% of the world average [2]. Therefore the energy consumption in China has been dominated by coal, which has accounted for more than 60% of total energy consumption. Since 1991, China has surpassed the United States to become the world's largest coal consumer. In 2017, coal consumption in China accounted for 60.4% of the total energy consumption. In addition, China's coal utilization is mainly based on power plant. And for a long time to come, the pattern of coal-fired power is difficult to change. In Chinese existing energy utilization system, coal accounts for two-thirds of the usage. While thermal power accounts for half of coal consumption in China. Therefore, the treatment of coal-fired flue gas has great significance for the environmental protection in China.

2. Air pollution from flue gas  
The serious smog weather generated since 2012 has made society pay more attention to air pollution. The cause of smog is mainly gaseous pollutants and fine particles discharged from fossil fuel combustion. More than 80% of Chinese annual coal consumption is direct combustion, which results in the largest amount of emissions of SO2, CO2, NOx, heavy metals and fine particulate matter in the world [3]. These pollutants can easily lead to air pollution, acid rain and greenhouse effect. SO2 and
NOx discharged into the atmosphere, part of which is combined with floating particles in the atmosphere, and combined with O₂ and metal oxides to generate aerosols by illumination. Then, a large amount of SO₂ and NOx are adsorbed to form fine particles, and dispersed in the air to form an acid mist. The other part is dissolved in rainwater through precipitation, which leads to the acid rain. The acidic ions will change the physicochemical properties of the soil, which will affect the roots of the plant, reduce crop yields and lead to acidification of water bodies, corrosion of metal and building [4, 5]. Air pollution has caused serious damage to the ecological environment and directly affects the health and survival of human beings around the world. Therefore, efficient and economical coal-fired flue gas disposal technology has significant meaning for atmospheric environmental protection, and will directly affect future social and economic development of China.

3. Flue gas desulfurization and denitration technology

At present, desulfurization and denitration are mainly divided into desulfurization technology and denitration technology. Desulfurization technology [6, 7] is divided into three categories according to the desulfurization time: pre-combustion, combustion and post-combustion flue gas desulfurization. Flue gas desulfurization (FGD) is the most widely used and mature technology to control SO₂ emission in thermal power plants. Since the middle of last century, the research on the control of coal-fired flue gas has developed rapidly. The conventional desulfurization methods in power plants can be divided into wet, dry and semi-dry methods according to the use of reagents and the shape of final products in the desulfurization process.

On the other hand, after decades of development, there are three main technical methods for NOx removal from flue gas: (1) denitration before combustion; (2) improvement of combustion and production process; (3) denitration of flue gas after combustion. Among them, denitration of flue gas after combustion is the most widely used way of NOx treatment at present [8]. A variety of flue gas denitration methods have been developed at home and abroad, such as selective catalytic reduction (SCR), non-selective catalytic reduction (NSCR), selective non-catalytic reduction (SNCR), catalytic oxidation, electron beam method (EBA), adsorption method and microbial method [9].

Next, two mature desulfurization and denitration technologies in industry will be introduced.

3.1. Desulphurization technology

The existing flue gas desulfurization technologies in China include limestone gypsum wet process, ammonia process, magnesium oxide process, semi-dry process, seawater desulfurization, double alkali process, etc. Limestone-gypsum wet method, semi-dry method, ammonia method and double alkali method are widely used, but seawater desulfurization is not widely used due to the limitation of conditions. According to the type difference of desulfurizer, flue gas desulfurization technology can be divided into dry method, wet method and semi-dry method. Among many processes, wet flue gas desulfurization (WFGD) technology is effective. It is the most used method, accounting for 85% of the total installed capacity of FGD units in the world.

3.1.1. Wet flue gas desulfurization

(1) Limestone/Lime-Gypsum Flue Gas Desulfurization

The maximum efficiency of wet limestone-gypsum desulfurization can reach 99%, which basically ensures that the SO₂ concentration in the flue gas can meet the requirements of China Air Pollutant Emission Standard for Thermal Power Plants. Therefore, this technology accounts for more than 85% of the existing devices in related industries. The process principle is as follows [10, 11]:

\[
2\text{CaCO}_3 + 2\text{SO}_2 + \text{O}_2 + 4\text{H}_2\text{O} \rightarrow 2\text{CaSO}_4 + 2\text{H}_2\text{O} + 2\text{CO}_2
\]  

(1)

Figure 1 is a typical limestone-gypsum desulfurization process. In the whole desulfurization process, the alkaline absorbent slurry reacts with flue gas in the spray tower, so that SO₂ contacts water and gets dilute acid, and then neutralizes with Ca(OH)₂ or CaCO₃ in the slurry to form calcium sulfite, which is refined and oxidized to high value-added gypsum. Although limestone-gypsum wet flue gas desulfurization has high desulfurization efficiency and extensive installed capacity, there are
also a series of problems [12, 13]. While desulfurizing, a large amount of CO₂ is released, which aggravates the pressure of national carbon emissions; the quality of desulfurized gypsum is poor, the cost of upgrading is high, there are a lot of problems of stacking, occupying space and polluting soil. Besides, in the process of equipment operation, water carrying capacity is large, with the extension of the whole process, a large amount of water resources are consumed.

Figure 1 Typical process of limestone-gypsum desulfurization

(2) Ammonia Flue Gas Desulfurization Technology

In recent years, with the increasing output of synthetic ammonia gas, ammonia flue gas desulfurization technology has developed rapidly and attracted widespread attention in China due to its high desulfurization rate, mature technology and low investment cost. Its technological principle is described as follows [14]:

\[
\begin{align*}
\text{SO}_2 + \text{NH}_3 + \text{H}_2\text{O} &= \text{NH}_4\text{HSO}_3 \\
\text{SO}_2 + 2\text{NH}_3 + \text{H}_2\text{O} &= (\text{NH}_4)_2\text{SO}_3 \\
\text{SO}_2 + (\text{NH}_4)_2\text{SO}_3 + \text{H}_2\text{O} &= 2\text{NH}_4\text{HSO}_3 \\
\text{NH}_3 + \text{NH}_4\text{HSO}_3 &= (\text{NH}_4)_2\text{SO}_3 \\
2(\text{NH}_4)_2\text{SO}_3 + \text{O}_2 &= 2(\text{NH}_4)_2\text{SO}_4
\end{align*}
\]

Ammonia water is used as desulfurizer in ammonia flue gas desulfurization technology. When the desulfurizer enters the desulfurization tower, it contacts and reacts fully with the flue gas containing SO₂, so as to remove SO₂. The primary product of ammonium sulfate was obtained, and the primary product will be oxidized to ammonium sulfate. After a series of refining processes, high value-added ammonium sulfate product [15] was obtained. Compared with the calcium method currently used in large-scale thermal power industry, ammonium method has obvious advantages. Ammonia has high activity, fast reaction speed, simple process, no fouling and clogging, easy to start and stop, easy to maintain, which is an environment-friendly technology without secondary pollution [16]. However, with the increasingly stringent environmental impact assessment, the old ammonium desulfurization equipment has been difficult to meet the desulfurization requirements, and the total dust of flue gas after desulfurization exceeds the standard.

(3) Dual-alkali flue gas desulfurization technology

In the wet desulfurization process, the scaling and blockage of equipment are easy to occur in the process site. In view of the above problems, a calcium-sodium double alkali process has been developed [17]. In the whole double alkali system, there are three main reactions:

① Desulfurization reaction

\[
\begin{align*}
\text{Na}_2\text{CO}_3 + \text{SO}_2 &\rightarrow \text{Na}_2\text{SO}_3 + \text{CO}_2 \\
2\text{NaOH} + \text{SO}_2 &\rightarrow \text{Na}_2\text{SO}_3 + \text{H}_2\text{O} \\
\text{Na}_2\text{SO}_3 + \text{SO}_2 + \text{H}_2\text{O} &\rightarrow 2\text{NaHSO}_3
\end{align*}
\]

② Regeneration reaction
\[
\text{Ca(OH}_2\text{)} + \text{Na}_2\text{SO}_3 \rightarrow 2\text{NaOH} + \text{CaSO}_3 \\
\text{Ca(OH}_2\text{)} + 2\text{NaHSO}_3 \rightarrow \text{Na}_2\text{SO}_3 + \text{CaSO}_3 + 2\text{H}_2\text{O}
\]

(10) (11)

Oxidation reaction

\[
\text{CaSO}_3 + \frac{1}{2} \text{O}_2 \rightarrow \text{CaSO}_4
\]

(12)

The technological process of double alkali flue gas desulfurization system is that the flue gas flows upward after entering the scrubbing tower, countercurrent and fully contacts with the spray liquid, and the SO\(_2\) gas in the flue gas is absorbed by the scrubbing liquid. The treated flue gas is dehydrated and emptied, and the absorbent liquid after reaction is discharged from the lower part of the cylinder, which is recycled by adding desulfurizer and by-product gypsum. The core advantage of double alkali desulfurization is that it has strong desulfurization ability and no clogging problem of wet desulfurization equipment. However, it has a large investment in the early stage and a large area and a high operating cost. In small-scale field applications, constrained by factors such as environment and capital, and its desulfurizer cannot be recycled, it actually increases the capital investment and environmental damage.

(4) Magnesium oxide wet flue gas desulfurization technology

The main chemical reaction processes of magnesium oxide wet flue gas desulfurization are as follows.

\[
\text{MgO} + \text{H}_2\text{O} \rightarrow \text{Mg(OH)}_2
\]

(13)

\[
\text{Mg(OH)}_2 + \text{SO}_2 + 5\text{H}_2\text{O} \rightarrow \text{MgSO}_3 \cdot 6\text{H}_2\text{O}
\]

(14)

\[
\text{MgSO}_3 + \text{SO}_2 + \text{H}_2\text{O} \rightarrow \text{Mg(HSO}_3)_2
\]

(15)

\[
\text{Mg(HSO}_3)_2 + \text{Mg(OH)}_2 + 10\text{H}_2\text{O} \rightarrow 2\text{MgSO}_3 \cdot 6\text{H}_2\text{O}
\]

(16)

The technological process is: (1) preparation of magnesium hydroxide emulsion; (2) the reaction of magnesium hydroxide emulsion and SO\(_2\) in the absorption tower; (3) the treatment of absorption liquid and sediment [18, 19]. The heated and pressurized flue gas is introduced into device and rapidly to cool down, and then the flue gas is washed by a quantity of magnesium hydroxide emulsion, and the flue gas is discharged after reaching the standard. Magnesium oxide wet flue gas desulfurization technology is an economical and efficient way of desulfurization. It has the characteristics of high desulfurization, low investment, small unit size and no environmental damage. However, the price of magnesia is high and its origin is concentrated.

3.1.2. Dry flue gas desulfurization technology

Dry flue gas desulfurization technology [20] means that the whole reaction is almost carried out in a dry state. Compared with wet flue gas desulfurization, the process has less investment and operation costs, and is easy to reprocess as a dry desulfurization product. There are some problems in this process, such as low desulfurization efficiency, large amount of absorbent in the unit, and loss of kinetic energy. At present, the technology of pulse discharge plasma flue gas desulfurization and microbial flue gas desulfurization are more mature.

3.1.3. Semi-dry flue gas desulfurization technology

Semi-dry flue gas desulfurization technology [21] generally includes spray drying desulfurization, circulating fluidized bed desulfurization, refluxing circulating fluidized bed flue gas desulfurization, new integrated desulfurization, gas suspension absorption, powder particle spouted bed, and furnace calcium injection desulfurization. Semi-dry flue gas desulfurization technology features that desulfurizer is sprayed into the flue gas in the form of solution and the absorbent reacts with SO\(_2\) in the form of gas liquefaction. At the same time, the water in the solution evaporates completely and the dry product is obtained. It has some advantages of both wet and dry desulfurization.

For example, in the spray drying process (SDA), lime slurry enters the absorber through the dynamic spray head, and the flue gas enters the absorber and contacts with the lime droplet. The residual heat of the flue gas removes the moisture in the lime slurry, and the flue gas is discharged after reaching the standard. Solid particles are collected and recycled by dust collectors. For new
integrated flue gas desulfurization (NID) technology, Ca(OH)₂ could be produced in a digester, then fluidized air is used as power, and the negative pressure in the pipeline is used to enter the reactor to contact the flue gas. In the process of contacting with flue gas, due to the large evaporation surface area of absorbent, moisture vaporization, temperature loss of flue gas and relative humidity increase, the flue gas after reaction needs to be dusted. Gas suspension absorption (GSA) method is that lime slurry enters from the bottom of the reaction tower through the two-fluid grouting system, and the flue gas enters from the bottom of the reaction tower as well. Solid particles are formed by reaction of atomized lime slurry with SO₂ in flue gas. The purified flue gas is discharged into the atmosphere after the dust collector. The main disadvantage is that the system is easy to scale and clog by using lime-eliminating emulsion as absorbent, and special equipment is needed to prepare absorbent, so the investment cost is too high, and the efficiency of desulfurization and the utilization rate of absorbent are not as high as that of limestone/gypsum method.

3.2. Denitration technology

Nitrogen oxides are the main cause of acid rain and photochemical smog. Nowadays, denitration in power plants mainly includes selective catalytic reduction and selective non-catalytic reduction.

3.2.1. Selective catalytic reduction

Selective catalytic reduction (SCR) [22] is a method of selective reduction of nitrogen oxides to nitrogen by using ammonia at 290-400℃ over catalysts, and ammonia hardly reacts with oxygen. In this way, the selectivity of ammonia is improved, and the consumption of ammonia could be decreased.

The main reactions are as follows:

\[
\begin{align*}
4\text{NH}_3 + 4\text{NO} + \text{O}_2 & = 4\text{N}_2 + 6\text{H}_2\text{O} \quad (17) \\
8\text{NH}_3 + 6\text{NO}_2 & = 7\text{N}_2 + 12\text{H}_2\text{O} \quad (18) \\
4\text{NH}_3 + 3\text{O}_2 & = 2\text{N}_2 + 6\text{H}_2\text{O} \quad (19) \\
4\text{NH}_3 + 5\text{O}_2 & = 4\text{NO} + 6\text{H}_2\text{O} \quad (20) \\
2\text{NH}_3 & \rightleftharpoons \text{N}_2 + 3\text{H}_2 \quad (21)
\end{align*}
\]

According to a large number of industrial practical experience, when the molar ratio of NH₃/NOₓ is near 1, the removal ratio of NOₓ can reach almost 90%, and the escape amount of ammonia in the second stage can be controlled below 5 mg/L. Due to the maturity of technology and high denitration ratio, SCR process has become the mainstream process of flue gas denitration for large industrial boilers worldwide. By the end of 2010, about 200 million kW flue gas denitration units had been put into operation in China, of which 95% were SCR units, which was 28% of the total capacity of coal-fired power units at that time.

3.2.2. Selective non-catalytic reduction

Selective non-catalytic reduction (SNCR) [23] refers to the reduction of nitrogen oxides in waste gas to H₂O and N₂ without the aid of catalysts. Nitrogen oxides are generally reduced by ammonia,
urea or hydrochloric acid as reductants in industry. The reductant reacts only with nitrogen oxides in flue gas, but generally does not react with oxygen. Because the process does not use catalysts, this method is called selective non-catalytic reduction. At the same time, due to the lack of catalyst, reducing agent must be added in the high temperature zone, generally in the furnace temperature range of 850-1100°C. SNCR is commonly used in boiler hearth, which could reduce NOX emission to about 200 mg/Nm3. On the basis of SNCR system in the furnace, a flue gas tail denitration device (SCR) was added to form the SNCR/SCR combined denitration process.

3.3. Combined desulfurization and denitration technology

At present, the process of step-by-step desulfurization and denitration technology is too complex, and its operation cost is often too high, so the combined removal technology of coal-fired flue gas has attracted more and more attention from researchers all over the world. Now the existing industrial combined removal technology is to use FGD in series with SCR, but this method does not complement each other. There are still some shortcomings of the original technology, such as easy scaling, blockage, corrosion and low economic efficiency. With the deepening of scientific research, a variety of combined removal technologies have gradually emerged, such as activated carbon absorption, flue gas circulating fluidized bed, chloric acid oxidation process, high-energy electron beam activation oxidation, low-temperature organic catalysis and so on.

1. Activated carbon absorption method

Activated carbon adsorption method [24] is to make the flue gas have suitable reaction conditions through the pretreatment of flue gas, and then enter the activated carbon absorption tower. The SO2 in the flue gas will be oxidized by the oxygen-containing complex group existing on the surface of activated carbon to form SO3, which reacts with water vapor to form H2SO4. For denitration, N2 is formed by oxidation-reduction reaction between NOX and NH3 under the condition of NH3. The disadvantage is that the operation cost is high and the quality of by-products is poor.

2. Flue Gas Circulating Fluidized Bed (CFB) Process

CFB technology [25] was originally a semi-dry desulfurization technology. After improvement, it has become a combined desulfurization and denitration technology. The principle is to use FeSO4·7H2O as catalyst, Ca(OH)$_2$ as the desulfurization agent and NH3 as the denitration agent. The by-product is CaSO4 and a small amount of CaSO3. And the double removal ratio of SO2 and NOX can reach more than 88%. However, CaCO$_3$ produced by desulfurization is very easy to cause environmental damage, and the efficiency of denitration will also decrease.

3. Chloric acid oxidation technology

Chloric acid oxidation technology [26] is a wet flue gas treatment process. The technology first oxidizes the flue gas by chloric acid, then absorbs it by alkali solution, so as to achieve the standard emission of flue gas. The technology is almost free from the restriction of external conditions and the double removal efficiency of SO2 and NOX is over 90%. However, chloric acid has strong corrosiveness and special requirements for equipment.

4. Electron beam activation technology

Electron beam activation (EBA) technique for flue gas purification [27] is an integration method of desulfurization and denitration, which uses electron gun to irradiate low temperature flue gas, ionize flue gas, then produce a series of ionic and free radical components used to oxidize SO2 and NOX in flue gas instantaneously, react with H2O and NH3 finally, and produce (NH4)2SO4 and NH4NO3 existing in solid state. At present, EBA method has gradually been industrialized and applied, which is one of the most potential combined desulfurization and denitration technologies. The advantages of EBA are dry treatment, no industrial waste, simple equipment, simple operation and easy control process. The double removal ratio of SO2 and NOX can reach more than 80%, and the by-products can be further processed as fertilizers. The disadvantage is the huge investment needed in the early stage.

5. Low-temperature organic catalysis

Low-temperature organic catalysis [28] is a new integrated process of desulfurization, denitration and dust removal at low temperature, which uses organic catalysts containing sulfoxide for
desulfurization and denitrification at low temperature. The reaction mechanism is that SO$_2$ in flue gas reacts with water to form sulfite, and covalent compounds are formed by combining sulfoxide with sulfite so as to inhibit the unstable sulfite decomposition. Because of the existence of oxygen in the flue gas, sulfite is continuously converted to sulfuric acid. The catalyst is then separated from sulfite, and the sulfuric acid is formed at the bottom of the tower with the introduced ammonia water to form ammonium sulfate fertilizer. The mechanism of denitrification is similar to that of desulfurization. Because NO is insoluble in water, it is necessary to convert NO into NO$_2$ or N$_2$O$_3$ dissolved in water by using strong oxidant O$_3$, and then nitrite is formed when it meets water. The effective component of organic catalyst, sulfoxide, is combined with nitrite to form a stable covalent compound, which makes nitrite unable to decompose. Because of the oxygen in the flue gas, nitrite is converted to nitric acid, and the catalyst could be separated from it. Ammonium nitrate fertilizer is produced by alkaline ammonia water and nitric acid. This technology is currently being used in the desulfurization and denitrification project of coke oven flue gas of a 1.2 million t/a coke scale coking plant in Shanxi Province.

4. Conclusion
Firstly, this paper introduces the current situation of energy structure and air pollution resulted from coal-fired in China, and clarifies that coal-fired industry is the main source of sulfur and nitrate pollutants. The emission control of coal-fired pollutants has become a serious problem faced by social and economic development and atmospheric environmental protection in China. The research progress of desulfurization and denitrification of coal-fired flue gas in China is reviewed. At present, the desulfurization and denitrification of coal-fired flue gas are generally conducted by limestone-gypsum method and selective catalytic reduction method. However, these two methods have many problems, such as complex process, large investment in the early stage, inefficient utilization of by-products, and cannot meet the current strict environmental protection requirements. Therefore, the economic and efficient integrated technology of desulfurization and denitrification will be the most promising development direction in the field of desulfurization and denitrification in the future.

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