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Coke Oven Flue Gas Desulfurization and Denitrification Technology Progress and Suggestions

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Abstract

With the development and progress of China's social economy, the people's quality of life is increasingly improved, people are increasingly concerned about environmental and ecological issues, and the relevant departments have higher and higher requirements for coke oven flue gas desulfurization and denitrification process technology. In the process of coke oven flue gas desulfurization and denitrification in coke plants, flue gas containing pollutants such as nitrogen oxides and sulfur dioxide will be emitted to the atmosphere, which will not only affect the environment, but also produce acid rain and cause serious pollution to the atmosphere. At present, the coke oven flue gas desulfurization and denitrification technology has been greatly improved, which has certain significance for the flue gas pollution management of coking enterprises. Based on this, this paper reviews the development status of coke oven flue gas desulfurization technology and puts forward relevant suggestions for the selection of desulfurization and denitrification process in coking industry.

Keywords

Coke oven flue gas; Desulfurization and denitrification technology; Application status; Development suggestions

1. Introduction

Coke oven flue gas (COFG) is the exhaust gas produced by the combustion of coke oven gas and air in the coke oven combustion chamber. It passes through the downdraft flue, chute area, regenerator, small flue, exhaust gas shutter, branch flue, main flue, desulfurization and denitrification equipment, and chimney before being discharged into the atmosphere.

Coke oven production processes are relatively unique, and its flue gas contains large amounts of nitrogen oxides and trace amounts of sulfur compounds, which pollute the atmospheric environment. Therefore, denitrification and desulfurization are essential before discharge. Based on actual conditions, the flue gas inside the coke oven has the following characteristics:

First, the flue gas temperature is relatively high, consistently maintaining a range of 210-320°C. Second, the composition of coke oven flue gas is relatively complex, and the sulfur dioxide content in coke oven flue gas is high, with an average concentration of 500 mg/Nm³, which increases the risk of pipeline corrosion. After desulfurization and denitrification of the coke oven flue gas, the flue gas temperature must be maintained above the dew point, and the denitrification system must be debugged to ensure its proper operation. In addition, in order to ensure the stable operation of the coke oven and the work safety and working environment of relevant employees, the relevant departments need to pay high attention.

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2. Analysis of the Progress of Desulfurization and Denitrification Technology of Coke Oven Flue Gas

2.1 Desulfurization Technology

2.1.1 Wet desulfurization technology

(1) Ammonia method

The principle of ammonia desulfurization technology is to allow ammonia to contact with sulfur dioxide in coke oven flue gas to produce a chemical reaction. This technology has high desulfurization efficiency and simple process flow. During the application process, there is no waste residue and no exhaust gas emission. However, at the same time, this technology also has certain defects, that is, ammonia escape often occurs during the application process, and the price of absorbent is relatively high, resulting in high desulfurization cost. In addition, during the application process of this technology, heavy metals, dioxins, etc. cannot be effectively removed [1].

(2) Lime/limestone method

This technology absorbs SO_2 in a wet scrubber through calcium oxide or calcium carbonate slurry. Compared with ammonia desulfurization technology, the desulfurization cost of lime/limestone method is lower and the desulfurization efficiency is as high as 95%. It is widely used because of its mature technology and stable operation. However, during the application process, there are also problems such as the difficulty of desulfurization wastewater treatment.

(3) Double alkali method

The so-called double alkali method refers to the process of treating SO_2, in which alkali metal sodium salt is first used to absorb SO_2, and then lime (limestone) is used to react with the generated Na_2SO_3 again. SO_2 is finally precipitated in the form of gypsum, and the alkali metal sodium salt can be recycled. The desulfurization efficiency of this method is over 90%, but due to the relatively complex process, the operating cost is relatively high, and the absorption liquid regeneration is difficult, which restricts the promotion and application of the double alkali method.

2.1.2 Semi-dry desulfurization technology

(1) Spray drying method (SDA)

This method mainly uses alkali solution, lime milk, limestone slurry, etc. as absorbents. Under the action of machinery or airflow, the absorbent is atomized, and then the flue gas is fully contacted and reacted to complete the desulfurization. During the application process, the efficiency of this technology can reach over 85%, the process is simple, and it will not cause serious corrosion to the equipment. However, the process has relatively high requirements for automation, and the amount of absorbent is difficult to control [2].

(2) Circulating fluidized bed (CFB) method

Based on the principle of circulating fluidized bed, it realizes the repeated recycling of absorbent, improves the utilization of absorbent and desulfurization efficiency. This method has low operating cost and will not cause corrosion, blockage and other problems to downstream equipment. However, the core technology and supporting equipment rely on imports, which limits the wide application of this process. Therefore, based on the actual domestic situation and local conditions, the development of circulating fluidized bed coke oven flue gas desulfurization technology that conforms to my country's national conditions and has independent intellectual property rights has become a hot research topic among domestic scholars. In addition, the content of calcium sulfite in the by-product of this process is higher than that of calcium sulfate. It must be operated close to the flue gas dew point to obtain a higher desulfurization rate, resulting in the enrichment of absorbent in the reactor. This is also where CFB desulfurization technology needs to be further improved [3].

2.1.3 Dry desulfurization technology

(1) In-furnace Calcium Injection Desulfurization Technology

In-furnace calcium injection desulfurization technology is a common commercial dry desulfurization process. The basic process involves injecting $CaCO_3$ or CaO powder into the boiler at temperatures between 800°C and 1150°C. The limestone powder has a particle size of approximately 1 mm or less, with a minimum particle size of no less than 120 μm. The bulk density ranges from 0.9 to 1.1 t/m³. The desulfurizer rapidly decomposes at high temperatures to produce CaO, which reacts with SO_2 in the flue gas to form $CaSO_3$, partially binding sulfur. However, in-furnace calcium injection desulfurization efficiency is often low, and desulfurizer utilization is also low.

(2) Baking Soda Dry Desulfurization (SDS)

This process uses baking soda as the desulfurizer. Sodium bicarbonate powder is injected into the desulfurization tower. Catalyzed by the high-temperature flue gas, sodium sulfate, carbon dioxide, and sodium thiocarbonate rapidly react with sulfur dioxide in the flue gas. After adsorption and purification, the treated products enter a dust collector for cleaning. The exhaust gas is then fed into a low-temperature SCR reactor, where nitrogen and water are generated with the help of a reducing agent. The process is relatively simple, with low initial investment and a small experimental site, and can be completed in a very short time. Using this process, the desulfurization rate is greater than 95%, and the denitrification efficiency is greater than 93%. The raw materials used are sodium bicarbonate and ammonia water, and the temperature is also relatively high [4]. The substances released are sodium sulfate and unqualified catalysts. The production of this method is highly safe, and maintenance and operation are relatively simple. The process does not produce wastewater or corrosive gases.

2.2 Denitrification Technology

(1) Low-nitrogen combustion technology

Low-nitrogen combustion technology is based on the NOx generation mechanism and changes the combustion conditions to achieve the purpose of controlling NOx generation. The main coke oven heating technologies include flue gas recirculation, staged heating, and actual combustion temperature control. Dust recovery is currently the most commonly used low-nitrogen combustion technology in the coking industry, and this technology is mostly used in existing coke ovens in China. Practice has shown that within the range of 10% to 20%, the appropriate control amount of flue gas recirculation is 10% to 20%. If it exceeds 30%, the combustion efficiency will decrease. When the coke oven is heated in stages, air and coal gas are usually used in staged heating to reduce its combustion intensity and achieve the purpose of reducing the amount of hot nitrogen oxides generated.

(2) Low-temperature SCR coke oven flue gas denitrification process

This technology is to pass a reducing agent such as ammonia, urea, etc. through a dedicated catalyst reactor at a certain temperature to cause the reducing agent to react with NOx in the flue gas to form nitrogen and water, thereby achieving denitrification. Low-temperature SCR flue gas desulfurization technology is a relatively mature and reliable denitrification technology among the current coke oven flue gas desulfurization technologies. It has high denitrification efficiency, simple and safe operation, and will not cause secondary pollution of large gases. The key to low-temperature SCR desulfurization technology lies in the catalytic effect of the catalyst, which can reduce the dependence on imported catalysts, prevent catalyst poisoning, and solve the secondary pollution of the catalyst [5].

3. Comparison of Coke Oven Flue Gas Desulfurization and Denitrification Technologies

The coke oven flue gas desulfurization and denitrification technologies currently used at home and abroad are: activated carbon dry desulfurization and denitrification, SDA semi-dry desulfurization + low-temperature SCR denitrification, and SDS dry desulfurization + low-temperature SCR denitrification. These three main process technologies are compared. The comparison of desulfurization and denitrification efficiency and process technologies is shown in Table 1, the comparison of material consumption and by-products is shown in Table 2, and the comparison of operating costs and operating stability is shown in Table 3.

Table 1. Desulfurization and denitrification efficiency and process technology comparison

	Process Specifications	Activated Carbon Dry Desulfurization and Denitrification	SDA Semi-Dry Desulfurization + Low-Temperature SCR Denitrification	SDS Dry Desulfurization + Low-Temperature SCR Denitrification
1	Desulfurization Efficiency	Available over 85%	Available over 80%-90%	Available over 95%
2	Denitrification Efficiency	Available over 70%	Available over 93%	Available over 93%
3	Desulfurizer	Activated Carbon	Fine Quicklime Powder	Finely Ground Industrial-Grade Sodium Bicarbonate
4	Denitrifier and Principle	Liquid Ammonia, Catalytic Reduction	Low-Temperature SCR Catalyst, Liquid Ammonia, Catalytic Reduction, Reaction Temperature 200-280°C	Low-Temperature SCR Catalyst, Liquid Ammonia, Catalytic Reduction, Reaction Temperature 180-240°C

Table 2. Material consumption and by-product comparison

	Process Specifications	Activated Carbon Dry Desulfurization and Denitrification	SDA Semi-Dry Desulfurization + Low-Temperature SCR Denitrification	SDS Dry Desulfurization + Low-Temperature SCR Denitrification
5	Water Consumption	None	Requires a certain amount of process water to be injected	None
6	By-Products	Dilute sulfuric acid, which can be used for sulfur resource recovery	Waste slag, primarily unstable $CaSO_3$	Sodium sulfate, which can be used for ceramic raw material recovery
7	Wastewater	None	None	None
8	Waste slag	None	Generates a large amount of dry waste slag, spent catalyst	Spent catalyst
9	Main Material Consumption	Activated carbon, liquid ammonia, electricity consumption	Quicklime powder, process water, electricity consumption, ammonia gas, catalyst replacement	Sodium bicarbonate, ammonia water, ammonia gas, electricity consumption, process water, catalyst replacement

Table 3. Operating Cost and Operational Stability Comparison

	Process Technical Specifications	**Activated Carbon Dry Desulfurization and Denitrification**	**SDA Semi-Dry Desulfurization + Low-Temperature SCR Denitrification**	**SDS Dry Desulfurization + Low-Temperature SCR Denitrification**
10	Operational Stability	Simple system, high flue gas emission temperature for dry desulfurization, no chimney corrosion protection required	Series process, complex system flow, prone to scaling and clogging, severe nozzle wear	Simple system, high flue gas emission temperature, no chimney corrosion protection required
11	One-time Investment	45-50 RMB/t coke production capacity	40-50 RMB/t coke production capacity	30-38 RMB/t coke production capacity
12	Operating Costs	10-13 RMB/t coke	10-12 RMB/t coke	4-8 RMB/t coke
13	Overall Evaluation	Fair	Fair	Excellent

A comprehensive evaluation of the above indicators shows that, in terms of the four key indicators of desulfurization efficiency, denitrification efficiency, byproduct disposal difficulty, and operating cost, SDS dry desulfurization combined with low-temperature SCR denitrification technology is most suitable for ultra-low emission retrofits of coke oven flue gas, and has already been successfully implemented in China.

4. Recommendations for the Development of Coke Oven Flue Gas Desulfurization and Denitrification Technologies

Currently, many domestic coke oven flue gas desulfurization and denitrification processes meet the relevant national special limit requirements. However, the process equipment used in this technology has high operating costs and requires significant investment. Furthermore, it will take time to verify its long-term stability and ability to meet emission requirements over the coking service life. Coke oven flue gas contains various impurities that can cause SCR catalysts to lose their activity. Therefore, relevant designers should take effective measures to optimize this process. At the same time, each production company should enhance its technical and operational capabilities and develop appropriate emergency response plans based on actual conditions to truly improve the quality and level of process equipment.

When selecting a process route, the appropriate one should be selected based on the flue gas conditions and site layout. In particular, the comprehensive utilization and digestion of desulfurization byproducts is a major challenge to prevent secondary pollution. Whether it is a new desulfurization and denitrification project, an existing desulfurization and denitrification device, or a project to be renovated, in order to ensure stable and efficient denitrification operation, it is recommended to prioritize the desulfurization-first, then the denitrification process.

5. Conclusion

In summary, with the continuous strengthening of my country's environmental protection policies, desulfurization and denitrification of coke oven flue gas have become a top priority. Enterprises should focus on strengthening control measures within the production process to achieve standard emissions. Enterprises in restricted areas, in particular, should consider adding dust treatment facilities to achieve standard emissions. To minimize ecological and environmental damage to industrial development, the application of science and technology must be strengthened. Pollutants such as SO_2 and NOx in coke oven flue gas are the main causes of environmental pollution. Therefore, in accordance with the characteristics of coke oven flue gas and relevant standards, increased efforts must be made to treat them. This article introduces the current status of coke oven flue gas desulfurization and provides an in-depth discussion of its technologies. To improve the overall quality of coke oven flue gas desulfurization and denitrification, it is necessary to increase investment in relevant areas and continuously improve and refine standard flue gas desulfurization and denitrification processes and technologies, laying a solid foundation for future desulfurization and denitrification efforts.

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