

# Optimization and reconstruction technology of SCR flue gas denitrification ultra low emission in coal fired power plant

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**Abstract.** In recent decades, nitrogen oxides (NO<sub>x</sub>) emissions from thermal power plant increased year by year in China. A large number of nitrogen oxides (NO<sub>x</sub>) emissions caused by the growing environmental problems have been widely attached importance to people. SCR denitrification technology has the advantages of cleanliness and high efficiency. At present, it has been the major technology to control NO<sub>x</sub> emission because of its high denitrification efficiency, reliable operation, no by-products and simple structure of the device. The denitrification efficiency can be stabilized at 70%. In this paper, three different denitrification methods are compared. The factors influencing the denitrification efficiency, the system arrangement and the key factors of the denitrification system are discussed in detail. And the numerical simulation of how to use this calculation software in the SCR reactor flue, baffle, reactor, spray ammonia grille and spray ammonia, mixer, etc. are reviewed, as well as the effect of system operation control on the deoxidation performance.

## 1. Introduction

With the rapid development of China's national economy, coal consumption is huge, accounting for about 70% of total energy consumption. China has a lack of oil and less gas, but coal resources are relatively rich, which is its basic national conditions. So, China's power industry is based on coal power generation. According to the statistics of nitrogen oxides (NO<sub>x</sub>) emissions in China's power industry in recent years ("National Environmental Bulletin (2014)"), there are 154633 industrial enterprises in China, the emission of nitrogen oxides (NO<sub>x</sub>) is 14.048 million tons. Among them, there are 3288 enterprises belonging to the thermal power industry, a total amount of 8.83 million tons of nitrogen oxides (NO<sub>x</sub>) are emitted, accounting for 55.62% of the total emissions. According to statistics released over the years, China's nitrogen oxides (NO<sub>x</sub>) emissions increased year by year, thermal power plant pollution problems become increasingly prominent, if it is not effectively controlled, will bring serious environmental damage.

Nitrogen oxides (NO<sub>x</sub>) is a highly toxic pollutant that can damage the ozone layer, the ecological environment and endanger human health. NO<sub>x</sub> is mainly generated by the coal, oil, natural gas and other petrochemical fuel combustion. NO<sub>x</sub> is composed by NO and NO<sub>2</sub>, which NO accounts for more than 90%. The affinity of NO<sub>x</sub> for hemoglobin is strong, once into the blood and hemoglobin firmly combines with the blood hypoxia, not only causing bronchitis, emphysema and other diseases, but also promoting premature aging, bronchial epithelial cell lymphoid tissue hyperplasia, and even lung cancer symptom. NO<sub>x</sub> emissions to the atmosphere will lead to the formation of O<sub>3</sub> and photochemical smog, resulting in human eye swelling, sore throat cough, skin flushing or even heart and lung failure and other symptoms. NO<sub>x</sub> with water will form HNO<sub>3</sub> and HNO<sub>2</sub>, resulting in formation of acid rain. NO<sub>x</sub> is also harmful to plants, causing leaf necrosis, whitening, yellowing or brown spots [1,2,3].



In order to protect the ecological environment, reduce NO<sub>x</sub> emissions, governments have developed a corresponding emission reduction regulations and standards. At present, the method of controlling the generation of NO<sub>x</sub> is mainly denitrification before combustion, denitrification in combustion and denitrification after combustion. Among the many denitrification technologies, the selective catalytic reduction (SCR) has many advantages such as no by-product, no secondary pollution, simple structure, reliable operation, easy maintenance and high denitrification efficiency, which has already been widely used for commercial applications. Meanwhile it is the most widely used flue gas denitrification technology in the international [4]. The early basic research of SCR denitrification technology is mainly focused on the reaction mechanism. Later, with the research of the reaction mechanism and the rapid development of computational fluid dynamics and computer technology, the numerical simulation technology has been widely used [5]. The use of CFD technology makes it possible to predict numerical values for complex flow, heat transfer, mixing and chemical reactions.

## 2. NO<sub>x</sub> control technology

In order to protect the ecological environment, the world has been committed to the development of advanced technology to reduce NO<sub>x</sub> emissions for a long time. According to the combustion life cycle, there are three categories to reduce NO<sub>x</sub> emission named pre-combustion denitrification, combustion denitrification and post-combustion denitrification respectively [6]. Pre-combustion denitrification is the way which low nitrogen fuel is used. But the engineering application is less because of the higher cost.

Generally, low NO<sub>x</sub> combustion technology is used in most combustion denitrification, which has lots of benefits like mature process, less investment and relative short construction period. But the denitrification efficiency is generally less than 40%. The countries like Japan and Germany, which have strict control on NO<sub>x</sub> emission always use the low NO<sub>x</sub> burner to reduce half of the NO<sub>x</sub> content first, then combine with post-combustion denitrification technology to further denitrify the flue gas [7,8,9].

Post-combustion denitrification can also be called flue gas denitrification technology and it can be divided into three categories, SCR, SNCR and SNCR/SCR mixed flue gas denitrification technology. It has already been used in Japan, the United States, Germany and other developed countries widely because of its high denitrification efficiency. SNCR technology is a process that the ammonia or urea reducing agent is sparged into the furnace followed by rapid decomposing of reducing agent and producing NH<sub>3</sub>, then NH<sub>3</sub> reacts with NO<sub>x</sub> to produce harmless N<sub>2</sub> and H<sub>2</sub>O which are the compositions of air. The operation temperature is about 1000°C with a 25%~50% denitrification efficiency. SCR denitrification technology is the means by adding a catalyst to reduce the activation energy of the reaction. The denitrification efficiency can reach about 95% at an operating temperature of about 400°C. The specific process of these two denitrification technologies is shown in Table 1. As for SNCR/SCR mixed flue gas denitrification technology, reducing agent is sparged into the furnace and then escaped ammonia and non-removal of NO<sub>x</sub> react as a catalytic reduction reaction to further remove NO<sub>x</sub>. The denitrification efficiency of this process can up to 40% to 80% [9,10].

By considering the economy strength of our enterprises and other elements, low-NO<sub>x</sub> combustion technology and flue gas denitrification technology are used together to remove NO<sub>x</sub> under current conditions of China. Environmental governance has long way to go. At the same time, China promotes the flue gas denitrification technology localization, industrialization vigorously, learns from foreign advanced experience and overcomes the NO<sub>x</sub> problem [11].

**Table 1** Comparison of SCR and SNCR Processes [8]

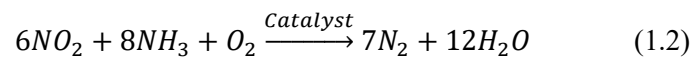
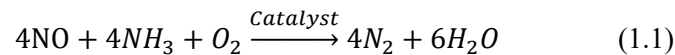
Process	SCR	SNCR
NO <sub>x</sub> removal efficiency/%	70~90	30~80
Operation temperature/°C	200~500	800~1100
Mole fraction of NO <sub>3</sub> /NO <sub>x</sub>	0.4~1.0	0.8~2.5
Leak of ammonia/10 <sup>-6</sup>	< 5	5~20
Total investment	High	Low
Operating costs	Medium	Medium

### 3. SCR technology

As more and more strict restriction has been putted on emission of NO<sub>x</sub> in fossil fuel power station of China, SCR becomes the primary technology used in China because of its low operating temperature, high NO<sub>x</sub> removal efficiency and no secondary pollution [10].

#### 3.1. Principle of SCR

The main components of NO<sub>x</sub> in the flue gas are NO and NO<sub>2</sub>. At low temperature, NO<sub>x</sub> reacts with the reducing agent NH<sub>3</sub> slowly. However, after catalyst has been introduced into reactor, the reaction can be carried out between 200°C and 450°C. The reaction equations are shown below:



In fuel gas, about 95% of the total amount of NO<sub>x</sub> is NO and NO<sub>2</sub> is only about 5%. So reaction equation (1.1) is the main reaction and also determines the reaction characteristics:

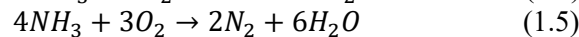
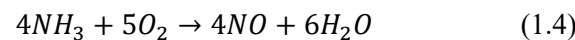
- (1) The mole ratio of NO to NH<sub>3</sub> is 1.
- (2) O<sub>2</sub> is required in this reaction.
- (3) The typical reaction temperature is between 300°C~400°C.

In addition to the above two main chemical reactions, denitrification reaction is also accompanied by the following side reactions.

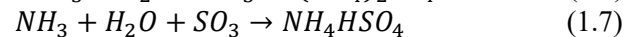
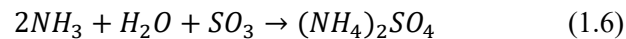
- (1) SO<sub>2</sub> oxidation reaction:



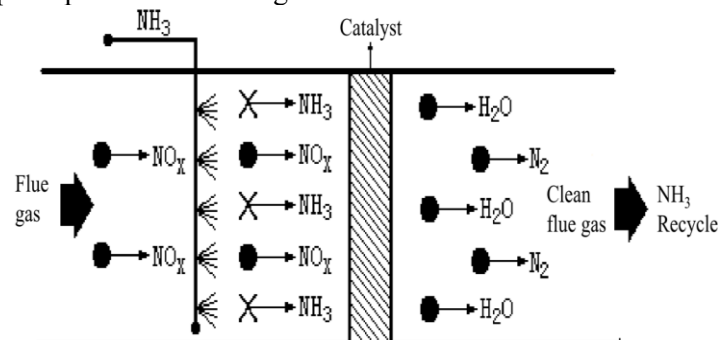
- (2) NH<sub>3</sub> oxidation reaction:



Ammonia and sulfuric acid reaction:



The fundamental principle is shown in Figure 1:



**Fig. 1** Schematic diagram shows the denitrification process of SCR

#### 3.2. SCR process parameters

- (1) SCR denitrification efficiency.

SCR denitrification efficiency is an important indicator of SCR denitrification effect, with the result affected by many factors. In engineering applications, the initial denitrification rate and the long denitrification rate are generally set. Now we can define  $\eta$  as denitrification efficiency:

$$\eta = \frac{(c_{in} - c_{out})}{c_{in}} \cdot 100\% \quad (2-1)$$

where:

$c_{in}$  = the concentration of  $\text{NO}_x$  before reaction.

$c_{out}$  = the concentration of  $\text{NO}_x$  after reaction.

### (2) SCR inlet $\text{NO}_x$ concentration

As the process of boiler combustion is more complex, the composition of coal is quite different, so the fluctuation range of the concentration of the inlet  $\text{NO}_x$  is relatively large and irregular.

### (3) Reaction temperature

Each catalyst has its own suitable temperature to play its catalytic activity. The reaction rate can only be ensured when its catalytic activity has been played fully. At present, SCR system always uses vanadium-titanium catalyst. On the one hand, if the reaction temperature is too low, the rate of reaction is quite low and side reactions may occur at the same time. On the other hand, if the reaction temperature is too high, the catalyst will be kneaded and is not good for the long-term use of the catalyst.

### (4) $\text{NH}_3/\text{NO}_x$ molar ratio

The definition of the molar ratio of  $\text{NO}_3/\text{NO}_x$  is:

$$a = \frac{c_{\text{NH}_3}^i}{c_{\text{NO}_x}^i} \quad (2-2)$$

where:

$c_{\text{NH}_3}^i$  = the inlet concentration of  $\text{NH}_3$  of the reactor.

$c_{\text{NO}_x}^i$  = the inlet concentration of  $\text{NO}_x$  of the reactor.

According to the characteristics of this reaction, usually  $\frac{n(\text{NH}_3)}{n(\text{NO}_x)} \approx 1$ . If the concentration of  $\text{NH}_3$  is too high, then the excess  $\text{NH}_3$  will enter the downstream area or the air, causing  $\text{NH}_3$  escape, neither economy, but also will cause downstream equipment corrosion, plug and other issues. Meanwhile, the equipment life will also be reduced. If the concentration of  $\text{NH}_3$  is too low, the requirements of efficiency of removing  $\text{NO}_x$  can not be reached. Therefore, a good SCR system needs to be supported with a good performance of the spray ammonia logic control system, spray ammonia grille, spray ammonia control valve to meet its molar ratio, and then achieve the efficiency requirements of  $\text{NO}_x$  off.

### (5) The escape rate of $\text{NH}_3$ .

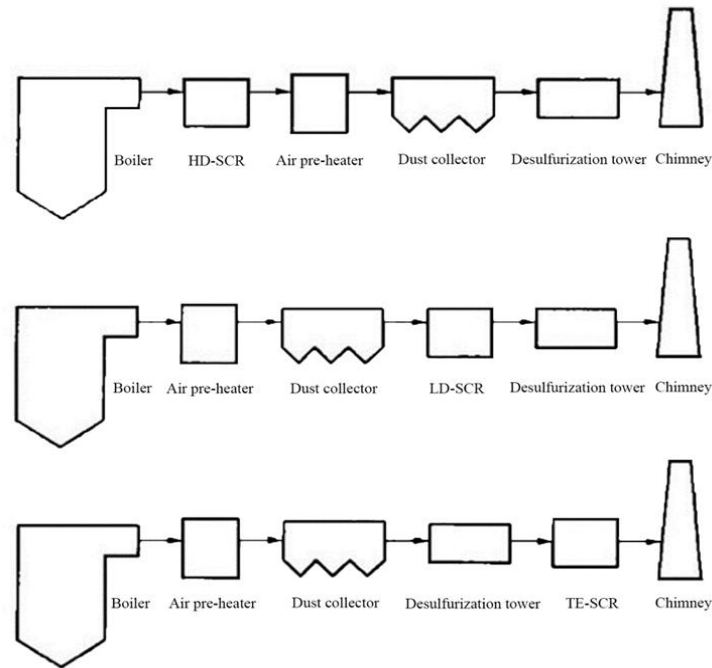
The  $\text{NH}_3$  escape rate refers to the volume fraction of unreacted  $\text{NH}_3$  in the flue gas to the total amount of the flue gas from  $\text{NO}_x$  outlet. If the  $\text{NH}_3$  escape rate is too high, not only increases in operating costs, but also causes secondary pollution. At the same time, the escape of  $\text{NH}_3$  will react with  $\text{SO}_3$ , which is the product of the oxidation of flue gas, to produce viscous  $\text{NH}_4\text{HSO}_4$ ,  $(\text{NH}_4)_2\text{SO}_4$  and other ammonium salts. It will cause the downstream equipment corrosion, reduce the service time of reactor. Normally, the permissible  $\text{NH}_3$  escape rate should not be greater than 3% [12].

### 3.3. SCR denitrification system layout

SCR denitrification system mainly consists of the reactor, liquid ammonia tank, ammonia/air mixer, sprayed ammonia grille, economizer and other components. According to the different position of the SCR reactor, there are three kinds of arrangements in the practical application: high temperature and high dust arrangement, high temperature and low dust arrangement, low temperature and low dust arrangement [8]. High temperature and high dust layout is commonly used in the current industrial. Three layouts are shown in Figure 2.

The first kind of layout is high temperature and high dust arrangement, the temperature of this kind of layout can meet the requirements of the reaction without additional heating, but the flue gas can

directly go into the reactor, a large number of fly ash which contains in the flue gas is easy to block the honeycomb catalyst, poisoning the catalyst by K, Na, Si and other plasma, reducing the catalyst life dramatically. The second kind of layout is high temperature and low dust arrangement. After the flue gas goes through an electrostatic precipitator, the disadvantage of the first layout can be avoided. But the operating reliability of the electrostatic precipitator at a temperature of 300°C~400°C is not high. The third one is low temperature and low dust layout. When the flue gas reaches the reactor, the temperature has dropped below 100°C. It means that extra heat is needed in order to meet the reaction requirements, resulting in waste of resources



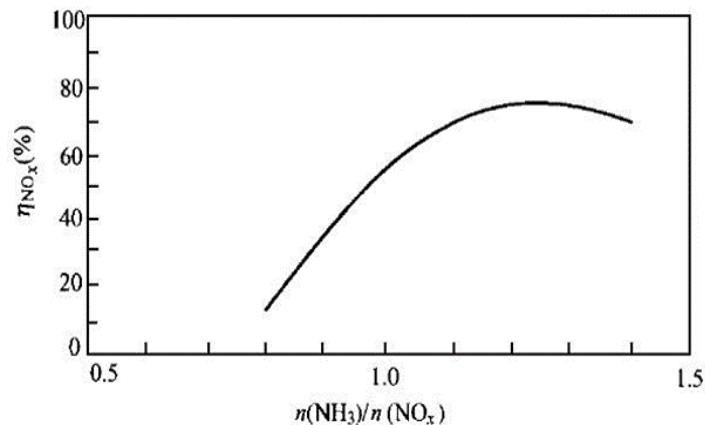
**Fig. 2** Three layouts of SCR

### 3.4. Key factors of SCR denitrification system

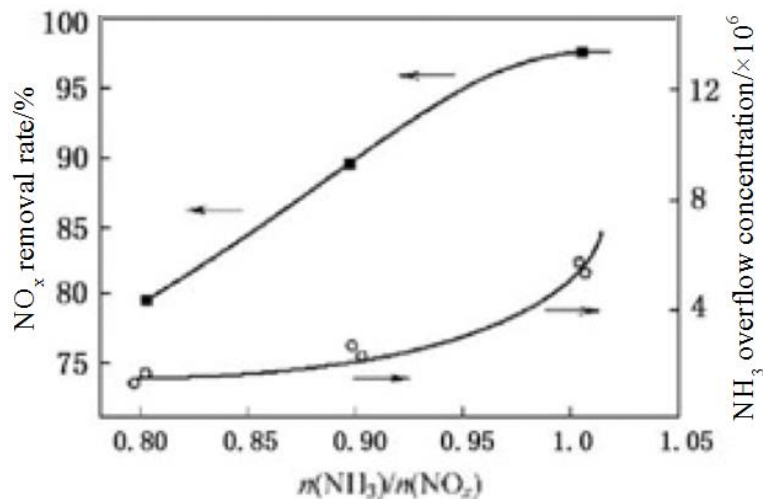
According to the characteristics, it can be seen that, there are many factors influencing the efficiency of NO<sub>x</sub> removal. The main factors are the reaction temperature, mixing uniformity of NO<sub>x</sub> and the reducing agent, catalyst performance and volume, inlet concentration of NO<sub>x</sub>, flowrate of flue gas, the molar ratio of ammonia over nitrogen and others [13,14,15].

If NO<sub>x</sub> in flue gas and reducing agent mixed uneven, it will cause the local concentration of reducing agent is too high or too low. If the local concentration of reducing agent is too high, then the excess NH<sub>3</sub> will enter the downstream area or air, resulting in NH<sub>3</sub> escape, neither economic, but also cause downstream equipment corrosion, plugging and other issues, reduce equipment life. If the local concentration of reducing agent is too low, it will reduce the removal efficiency of NO<sub>x</sub>, fail to meet the design requirements.

It can be seen from Figure 3 and Figure 4, when  $n(\text{NH}_3)/n(\text{NO}_x) < 1.25$ , the removal efficiency of NO<sub>x</sub> increases with the increase of  $n(\text{NH}_3)/n(\text{NO}_x)$ . But according to the results of the research and the synthesis of various factors, the removal NO<sub>x</sub> efficiency can reach more than 95% and the escape rate of ammonia is low, which is the optimal result of SCR design [16].



**Fig. 3** The relationship between the denitrification efficiency and the molar ratio of  $\text{NH}_3/\text{NO}_x$



**Fig. 4** The relationship between SCR denitrification efficiency and ammonia escape rate as well as  $\text{NH}_3/\text{NO}_x$  molar ratio

#### 4. Application of numerical simulation in scr technology

Computational Fluid Dynamics (CFD) is a professional software based on discrete numerical calculation method to simulate numbers and analyze the phenomena of fluid flow as well as heat transfer. At present, there are three methods to study the problems of fluid mechanics: experimental analysis, theoretical analysis, CFD simulation. Although the results of the experimental analysis are relevant reliable, the experimental measurement takes longer and higher cost is required. Theoretical analysis is difficult to explain the complex flow mechanism of turbulence. But CFD simulation, which abandon the shortcomings of experimental analysis and theoretical analysis, saves time and effort, is an efficient solution to a variety of flow problems [17,18].

Generally, there are three steps for CFD to solve the problem: pre-processing, calculation, post-processing. Gambit is a commonly used pre-processing software that can generate tetrahedron, hexahedral, pyramid & prismatic structured and unstructured meshes. And for complex geometry, Gambit software can partition it and within the partition, high quality grids can be generated. Fluent software is a commonly used fluid mechanics calculation software with a higher accuracy, which can adopt the rich physical model such as structural grid, instructed grid and mixed grid. The algorithm is based on the uncoupled implicit algorithm and coupled display/implicit algorithm etc. After a long period of development, the NITA algorithm greatly shortens the calculation time. The model, which has already processed by the Gambit preprocessing, can be directly imported into the Fluent software, and the corresponding parameters can be set up to calculate the solution, and its own has post-processing



function. Tecplot is a post-processing software with drawing, data analysis and powerful data display function. With its continuous improvement, it can be widely used in more and more engineering research [19].

With the development and the perfection of Fluent software, more and more chinese and foreign researchers have used Fluent software to optimize the flue, the baffles, the rector, the sprayed ammonia barrier of the SCR system and the amount of ammonia spray, the mixer and so on. Good results have been achieved.

### 5. Effects of SCR System operation control on removal NO<sub>x</sub>

It has been pointed out that SCR denitrification technology has the optimal result of SCR design when  $n(\text{NH}_3)/n(\text{NO}_x) \approx 1$ . So,  $n(\text{NH}_3)$  and  $n(\text{NO}_x)$  need a logic control system to control its ratio. At present, in the construction and transformation of Chinese coal-fired power plants, although the automatic control system can basically complete the design requirements, poor control accuracy, lag of adjustment and other issues will occur under some specific conditions. Due to different coal composition and different combustion process, the flowrate of reactor inlet flue gas and NO<sub>x</sub> concentration fluctuations are relatively large and irregular. If a large amount of ammonia is needed, the valve can be adjusted to meet the accuracy requirements. But if tiny amount of ammonia is needed, due to the limitation of the mechanical properties of the valve, when the operating point downs to a certain extent, the adjustment accuracy cannot meet the requirements. At the same time, the data of CEMS measurement is unstable when the denitrification flue gas continuous detection system (CEMS) is periodically cleaned, resulting in the hysteresis of the sprayed NH<sub>3</sub> regulation. And due to the instability of sprayed ammonia, a significant reduction in the efficiency of removal NO<sub>x</sub> and a much higher escape rate of ammonia can be seen. The operating life of device can also be decreased. Therefore, in order to ensure the efficiency of removal NO<sub>x</sub> and enhance the continuity, reliability and economy of SCR denitrification system, it is necessary to optimize the matching automatic control system [20,21].

### 6. Conclusion

With the progress of society, the demand for electricity has increased constantly. But the contradiction between environmental pollution and the demand of electricity is growing. Under pressure from the environment, in China, the emission standards for NO<sub>x</sub> of the coal-fired power plant is almost harsh. SCR denitrification technology has been popularized. In this context, it is imminent to optimize its equipment structure, from the perspective of thermodynamics to improve its process parameters, explore the feasibility of the proposed program and find its laws. Meanwhile, the most important work of the current is to optimize the existing equipment.

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