Low-Temperature SCR Catalyst Development

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In recent years, low-temperature SCR (Selective Catalytic Reduction) denitrification technology has been popularized in non-power industries and has played an important role in the control of industrial flue gas NOx emissions in China. Currently, the most commonly used catalysts in industry are V2O5-WO3(MoO3)/TiO2, MnO2-based catalysts, CeO2-based catalysts, MnO2-CeO2 catalysts and zeolite SCR catalysts. The flue gas emitted during industrial combustion usually contains SO2, moisture and alkali metals, which can affect the service life of SCR catalysts.

Keywords: selective catalytic reduction; nitrogen oxides; low temperature

1. Introduction

Nitrogen oxides (NO_x) , including NO and NO₂, are considered as the main air pollutants from industrial and automobile exhausts, which have caused a lot of environmental problems, such as haze issues and ozone depletion. Until now, the selective catalytic reduction of NO_x by ammonia $(NH_3\text{-SCR})$ is accepted to be an effective method to eliminate NO_x pollutants $\frac{[1]}{2}$.

In SCR denitration technology, the most important thing is the catalyst that should possess the high activity, excellent sulfur oxides and water resistance abilities. The traditional V-based catalysts showed good deNO_x performance at 300–420 °C, which have been well used to abate the flue gas from the power plants. Due to the wide applications of SCR technology, the NO_x emissions from power industry have been well controlled, while NO_x emission control in the non-power industry faces severe challenges due to the low temperature of the flue gas, which is usually below 300 °C. Therefore, it is difficult to use traditional SCR catalysts for the gas pollutant treatment in the non-power industry. In the past ten years, Chinese scholars and industries have made great efforts on denitration in the non-power industry and made remarkable achievements. Wang and Dong et al. [1][2] present the detailed information concerning NH₃-SCR in some non-power industries, showing that the gas condition was more complex and fluctuated than that from the power plant.

2. Low-Temperature SCR Process

Usually, the flue gas temperature of the industrial process, such as in coking and steel sintering processes, is lower than 300 °C and contains many constituents with low concentrations, such as sulfur dioxide and alkali metal salts, and large amounts of water are also present. Thus, the SCR catalyst must be active in the low-temperature range (typically in the range of 160-300 °C) and stable under harsh gas conditions with good sulfur oxides and water resistance performance $^{[3]}$. In the typical flue gas treatment system of power plants in China, which usually employ high-dust SCR system to control NO_X emissions, the SCR reactor is located upstream of the particulate control devices and flue gas desulfurization (FGD) system. The so-called "medium- and high-temperature SCR" units can be operated in the temperature range of 280-400 °C. For NO_X emission control in the non-power industry, especially in the coking and steel sintering industry, the "low-dust" or "tail-end" configuration of SCR technology should be adopted to decrease the impacts of SO_2 and dust on the SCR catalyst. The low-temperature SCR reactor is located downstream of the particulate control devices and flue gas desulfurization (FGD) system. The SO_2 concentration and dust amount in inlet of the SCR reactor should be lower than 35 and 5 mg/m 3 , respectively, to meet the ultra-low emission standards. In the key areas of coal-fired boilers, the value should be lower than 50 and 20 mg/m 3 . In this case, the operating temperature of the SCR unit can be decreased to 160 °C. Although SO_2 and dust in the flue gas have very low values, the SCR catalyst needs to operate for three years at a low temperature, posing a severe challenge to SCR catalyst technology.

3. SCR Catalysts

Low-temperature SCR technology is an economic and effective process in abating the NO_x pollutants emitted from the non-power industry. Based on the consensus of the advantages that low-temperature SCR technology possesses, in the last decade, considerable research in China have been devoted to developing catalysts that can work well under low-temperature conditions. The development and research in SCR catalysts have been reviewed and summarized [2][3]. As reported, many kinds of low-temperature SCR catalyst system have been proposed and investigated. The main low-temperature SCR catalyst systems include V_2O_5 -WO₃(MoO₃)/TiO₂, Mn complex oxides, CeO₂-based and zeolite catalysts.

3.1. V₂O₅-WO₃(MoO₃)/TiO₂

As a typical and efficient catalyst, a V_2O_5 - $WO_3(MoO_3)/TiO_2$ catalyst used in NH₃-SCR technology has been commercialized for several decades. The typical commercial catalyst used in power stations presents low activity at low temperatures (below 300 °C) and cannot meet the need to abate NO_x from non-power industries.

In the past decades, various transition-metal oxides have been researched as catalysts for NH_3 -SCR at low temperatures. In order to meet the need of activity, stability, and resistance of SO_2 and H_2O , plenty of methods have been tried to improve SCR performance.

The most direct and convenient method to improve the low temperature activity of V_2O_5 -WO₃(MoO₃)/TiO₂ catalyst is to appropriately increase the loading of V_2O_5 . However, when V_2O_5 loading increases, the oxidative ability of the catalyst will be increased leading to the enhancement of SO₂ conversion. This is not allowed for the low-temperature SCR technology. Therefore, the catalyst needs to coordinate the redox activity and the surface acid property, reduce the adsorption of SO₂ on the catalyst surface and suppress the oxidation of SO₂. In another way, the NH₃ adsorbed on the Lewis acid sites (V^{5+} -O) on V_2O_5 -WO₃(MoO₃)/TiO₂ catalysts can react with NO at low temperatures [4]. By increasing the surface acidity and inhibiting the oxidative ability over the SCR catalysts, the operating temperature window of the V_2O_5 -MoO₃/TiO₂ catalyst is expanded to the range of 160–400 °C, which also shows acceptable SO₂ and H₂O resistance at low temperatures [5][6][7]. The V_2O_5 -MoO₃/TiO₂ catalysts have been used intensively in denitration reaction projects of coking sintering, refuse incinerators and other non-electric industries in China.

Another way to improve the low-temperature SCR activity of the V_2O_5 -WO₃(MoO₃)/TiO₂ catalyst is through modification and doping by introducing other elements into the catalyst system, which is easy to achieve in practice due to the convenience in the preparation. For example, Zhang et al. [8] investigated the effect of Mn, Cu, Sb, and La doping on the SCR performance of the V_2O_5 -WO₃(MoO₃)/TiO₂ catalyst. The investigation showed that Mn and Cu could enhance the redox property and weak surface acidity, while Sb and La addition showed promotion in the amount of acid sites. Liang et al. [9] demonstrated that a 3% addition of CeO₂ improved the NH₃ adsorption performance, NO oxidation, and sulfur oxide and the water-resistance of the V_2O_5 -WO₃/TiO₂ catalyst.

3.2. MnO₂-Based Catalysts

Manganese-containing catalysts have been paid enough attention due to their variable valence states and excellent redox properties. However, for its poor N_2 selectivity and easy deactivation by SO_2 , the catalyst only containing manganese oxide is extremely restricted in the SCR process. Mn-based composite oxides are popular and have proven to be effective catalysts with an enhanced SCR performance [10].

Over MnO_x catalyst, NH₃ species adsorbed on Lewis acid sites (Mn³⁺) were active at low temperatures. Bidentate nitrates were inactive at low temperatures (below ~225 °C), but active at higher temperatures $^{[11]}$.

Mn-based composite oxides possess excellent redox properties due to their various valence state, which are a benefit to enhance the process of NO oxidation to NO₂. The formation of NO₂ from NO oxidation is considered as a key factor in low-temperature activities because a certain concentration of NO₂ gives an enhancement of the "Fast SCR" reaction at low temperatures. Chen et al. $\frac{[12]}{}$ proposed that the redox cycle between $Cr^{5+} + 2Mn^{3+} \leftrightarrow Cr^{3+} + 2Mn^{4+}$ promoted the oxidization of NO to NO₂ at low temperatures. Liu et al. $\frac{[13]}{}$ reported that an urchin-like MnCrO_x catalyst possessed good NH₃-SCR activity in the temperature range of 150–350 °C and improved SO₂ resistance.

Gao et al. $\frac{[14]}{}$ discovered that CoMnO_x showed high NH₃-SCR activity at low temperatures and delayed the trend of SO₂ poisoning. Zhao et al. $\frac{[15]}{}$ found that a lamellar CoMnO_x composite oxide could provide more Lewis acid sites and surface oxygen species than those of CoMnO_x nanoparticles. Wang et al. $\frac{[16]}{}$ reported that ballflower-like CoMnO_x catalyst

exhibited good SCR activity and N_2 selectivity in the temperature range of 150–350 °C, showing a certain amount of SO_2 resistance and durability.

The doped component was usually considered to give a promotion of surface lattice oxygen species. Sun et al. $^{[17]}$ investigated the NH₃-SCR activity over the Nb-doped Mn/TiO₂ catalysts with the optimum Nb/Mn molar ratio of 0.12. Rare earth elements were also used in the modifications. Liu et al. $^{[18]}$ and Xu et al. $^{[19]}$ investigated the effect of the introduction of Sm to Mn-TiO_x catalysts. The introduced Sm could improve the dispersion of manganese oxide on the surface of the catalysts, resulting in increases in surface area, the amount of weak Lewis acid sites and surface oxygen.

Among these catalysts, spinel-type materials containing manganese attracted interest in SCR due to their special spatial structures and physical-chemical properties. Gao et al. $^{[20]}$ reported that a Zr^{4+} cation doped $MnCr_2O_4$ spinel, the zirconium incorporated in the crystal of $MnCr_2O_4$ produced higher levels of beneficial Mn^{3+} , Mn^{4+} and Cr^{5+} species, and showed an increase in the acidity and redox ability.

However, these catalysts are very sensitive and exhibit unsatisfactory N_2 selectivity [21]. The stability in the presence of SO_2 and H_2O in the flue gas is still a problem for MnO_2 -based catalysts.

3.3. CeO₂-Based Catalysts

He et al. $^{[22][23]}$ reported that the crystal structure, crystallite size and catalytic NH₃-SCR activity over the CeO₂-based catalysts presented a regular change with the increase in CeO₂ concentration. Particularly, the CeO₂-TiO₂ (1:1 in weight) catalyst with an amorphous structure showed a higher BET surface area and a stronger surface acidity than other samples. Meanwhile, favorable Ce³⁺ and the surface-adsorbed oxygen benefited the adsorption of NO_x and NH₃ molecules, which could enhance NH₃-SCR activity.

In the past years, tungsten or molybdenum addition in ceria-based catalysts was paid some attention. Jiang et al. $^{[24]}$ demonstrated that the introduction of WO₃ could improve SCR activity over the CeWTiO_x catalysts due to the enhanced dispersion of Ce species over TiO₂ and the amount of Ce³⁺ and chemisorbed oxygen. Li et al. $^{[25]}$ investigated the adsorption and reactivity of NH₃ and NO over the CeWTiO_x catalyst, showing that over 90% of NO conversion can be obtained in the temperature range of 250–500 °C. Liu et al. $^{[26]}$ reported that the WO₃/TiO₂@CeO₂ core–shell catalyst present a synergistic effect of redox properties and acidity, which is in favor of the excellent NH₃-SCR activity and better SO₂ resistance. Kim et al. $^{[27]}$ found monomeric W in CeO₂/TiO₂ catalyst enhance the SCR reaction activity at low temperatures due to the increased NO adsorption and the formation of unstable NO_x adsorption species. Dong et al. $^{[28]}$ presented that the coverage of MoO₃ weakened the adsorption of nitrate species over the CeO₂-TiO₂ catalyst, giving an increase in the number of Brønsted acid sites. For the CeO₂-based catalyst, cerium sulfate, which is formed in reaction with SO₂ in the flue gas during the SCR process at high temperatures, has attracted wide attention. Fan et al. $^{[29]}$ showed that NH₃-SCR reaction over CeO₂/TiO₂-ZrO₂-SO₄²⁻ mainly followed the L-H mechanism at a low temperature (250 °C) and the E-R mechanism at 350 °C.

Generally, for the limitation of sulfate formation and low temperature SCR performance, CeO₂-based SCR catalyst does not seem an optimal choice for NO_x elimination under low-temperature conditions at present.

3.4. MnO₂-CeO₂ Catalysts

Rare-earth metal oxides, such as Ce, have been frequently adopted to modify MnO_x as an efficient low-temperature NH_3 -SCR catalyst due to their incomplete 4f and empty 5d orbitals $\frac{[30]}{100}$. Leng et al. $\frac{[31]}{100}$ synthesized $MnCeTiO_x$ catalysts and compared the NH_3 -SCR activity over the samples with different Mn/Ce mol ratios in the low-temperature range. The results showed that the low-temperature SCR activity over $MnCeTiO_x$ compositions was greatly improved due to the incorporation of Mn, and the best performance (~100% NO conversion and above 90% N_2 yields) across the temperature range of 175–400 °C at GHSV of 80,000 h^{-1} .

In the published lectures, composite transition metal oxides usually showed a higher activity than single oxide materials. Zhang et al. $^{[32]}$ demonstrated the enhanced electron mobility effect that originated from MnO_x and CeO_x , which enhanced low-temperature de NO_x efficiency. Compared to the single composition of CeO_2 , MnO_x could increase the pore volume and pore diameter, and enhance the adsorption of NO and NH_3 as well as in the concentrations of Ce^{3+} on the CeO_2 - MnO_x catalyst, which is beneficial to increase the redox properties $^{[33]}$.

Yang et al. [34] studied SCR activity over the activated carbon supported Mn-Ce oxide catalysts modified by Fe, on which ca. 90% NO conversion was obtained at 125 °C with GHSV of 12,000 h⁻¹. Zhu et al. [35] synthesized a 3D-structured MnOx-CeO₂/reduced graphene oxide, giving a NO_x conversion of 99% at 220 °C with GHSV of 30,000 h⁻¹.

3.5. Zeolite SCR Catalysts

lon-exchanged zeolite catalysts with small pores have been accepted as optimum SCR catalysts in NO_x elimination from diesel engine exhaust. Among them, copper or iron exchanged zeolites with a chabazite (CHA) structure, such as Cu/SAPO-34 and Cu/SSZ-13, have received significant attention due to their excellent SCR performance, wide temperature window and thermal stability in harsh conditions [36]. Cu-SSZ-13 exhibits excellent SCR activity (>80% NO conversion) and N_2 selectivity in a wide temperature range of 150–450 °C [37]. Cu/SAPO-34 prepared by a hard-template method using CaCO₃ as template present NO_x convention above 90% at 170–480 °C, even introducing 10% H_2O [38]. A heterobimetallic FeCu-SSZ-13 zeolite with high crystallinity was prepared by an economic and sustainable one-pot synthesis strategy, which presents a wide reaction temperature window, excellent hydrothermal stability, high H_2O and SO_2 tolerance, and good gaseous hourly space velocity flexibility [39].

Zeolite catalysts for SCR has been developed rapidly these years, offering a great contribution to abate the NO_x reduction. However, they may be not the optimum choice for NO_x elimination in stationary sources due to the limitations of cost and synthesis strategy.

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