

Review

Low-Temperature SCR Catalyst Development and Industrial Applications in China

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Abstract: 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 NO_x 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. This paper summarizes the mechanism of catalyst poisoning and aims to reduce the negative effect of NH₄HSO₄ on the activity of the SCR catalyst at low temperatures in industrial applications. It also presents the outstanding achievements of domestic companies in denitrification in the non-power industry in recent years. Much progress has been made in the research and application of low-temperature NH₃-SCR, and with the renewed demand for deeper NO_x treatments, new technologies with lower energy consumption and more functions need to be developed.

Keywords: selective catalytic reduction; nitrogen oxides; low temperature; catalyst poisoning; non-power



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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 (NH₃-SCR) is accepted to be an effective method to eliminate NO_x pollutants [1].

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. This review focus on the research and development in low-temperature SCR and its application in non-power industry, especially on the anti-sulfur and water resistance properties of the catalyst, as well as on how to reduce the negative impact of

ammonium bisulfate (ABS, NH_4HSO_4) on the catalyst activity at low temperatures in industrial applications.

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 $300\text{ }^\circ\text{C}$ 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 $160\text{--}300\text{ }^\circ\text{C}$) 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 $280\text{--}400\text{ }^\circ\text{C}$. 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 $160\text{ }^\circ\text{C}$. 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 $\text{V}_2\text{O}_5\text{--WO}_3(\text{MoO}_3)/\text{TiO}_2$, Mn complex oxides, CeO_2 -based and zeolite catalysts.

3.1. $\text{V}_2\text{O}_5\text{--WO}_3(\text{MoO}_3)/\text{TiO}_2$

As a typical and efficient catalyst, a $\text{V}_2\text{O}_5\text{--WO}_3(\text{MoO}_3)/\text{TiO}_2$ catalyst used in NH_3 -SCR technology has been commercialized for several decades. The typical commercial catalyst used in power stations presents low activity at low temperatures (below $300\text{ }^\circ\text{C}$) 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 $\text{V}_2\text{O}_5\text{--WO}_3(\text{MoO}_3)/\text{TiO}_2$ 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 SO_2 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 SO_2 on the catalyst surface and suppress the oxidation of SO_2 . In another way, the NH_3 adsorbed on the Lewis acid sites ($\text{V}^{5+}\text{--O}$) on $\text{V}_2\text{O}_5\text{--WO}_3(\text{MoO}_3)/\text{TiO}_2$ 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 $\text{V}_2\text{O}_5\text{--MoO}_3/\text{TiO}_2$ catalyst is expanded to the range of $160\text{--}400\text{ }^\circ\text{C}$, which also shows acceptable SO_2 and H_2O resistance at low temperatures [5–7].

The V_2O_5 - MoO_3 / TiO_2 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 - $WO_3(MoO_3)$ / TiO_2 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 - $WO_3(MoO_3)$ / TiO_2 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 CeO_2 improved the NH_3 adsorption performance, NO oxidation, and sulfur oxide and the water-resistance of the V_2O_5 - WO_3 / TiO_2 catalyst.

3.2. MnO_2 -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, NH_3 species adsorbed on Lewis acid sites (Mn^{3+}) 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 NO_2 . The formation of NO_2 from NO oxidation is considered as a key factor in low-temperature activities because a certain concentration of NO_2 gives an enhancement of the “Fast SCR” reaction at low temperatures. Chen et al. [12] proposed that the redox cycle between $Cr^{5+} + 2Mn^{3+} \leftrightarrow Cr^{3+} + 2Mn^{4+}$ promoted the oxidization of NO to NO_2 at low temperatures. Liu et al. [13] reported that an urchin-like $MnCrO_x$ catalyst possessed good NH_3 -SCR activity in the temperature range of 150–350 °C and improved SO_2 resistance.

Gao et al. [14] discovered that $CoMnO_x$ showed high NH_3 -SCR activity at low temperatures and delayed the trend of SO_2 poisoning. Zhao et al. [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. [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 NH_3 -SCR activity over the Nb-doped Mn/ TiO_2 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. CeO_2 -Based Catalysts

He et al. [22,23] reported that the crystal structure, crystallite size and catalytic NH_3 -SCR activity over the CeO_2 -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 NH₃-SCR catalyst due to their incomplete 4f and empty 5d orbitals [30]. Leng et al. [31] synthesized MnCeTiO_x catalysts and compared the NH₃-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% N₂ 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 deNO_x efficiency. Compared to the single composition of CeO₂, MnO_x could increase the pore volume and pore diameter, and enhance the adsorption of NO and NH₃ as well as in the concentrations of Ce³⁺ on the CeO₂-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 MnO_x-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

Ion-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 N₂ selectivity in a wide temperature range of 150–450 °C [37]. Cu/SAPO-34 prepared by a hard-template method using CaCO₃ as tem-

plate present NO_x conversion 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.

4. Deactivation of Low Temperature SCR Catalyst

The flue gas emitted from industrial combustion processes usually contains SO_2 , moisture, alkali metals, among others. These factors can reduce the activity of catalysts. Figuring out the mechanism of poisoning and how to reduce the effect of these factors has been a problem faced by the researchers as there are many reports discussing this issue.

4.1. SO_2 Poisoning

A primary mechanism of SO_2 deactivation over the catalyst at low temperatures can be observed in three ways. (i) While SO_2 can be oxidized to SO_3 , SO_3 can further react with NH_3 and H_2O to form $(\text{NH}_4)_2\text{SO}_4$ and NH_4HSO_4 to cover the catalyst surface, resulting in the reduction in surface area, pore volume and pore size of the catalyst, and causing reversible activity loss. (ii) The SO_2 reacts with the active components (mainly transition metals) on the catalyst surface to form metal sulfates and induce a more severe and irreversible loss of activity. (iii) SO_2 showed a strong competitive adsorption effect to NO on the catalyst surface, which would reduce the formation of SCR intermediates and its catalytic efficiency, in order to improve the sulfur resistance of low-temperature SCR catalyst, which can be mainly taken in the following four ways.

4.1.1. Reduction in SO_2 Adsorption/Oxidation

The addition of elements, such as Co, Sn, Cu, V, Cr, Fe, as additives to the catalyst, can create a synergistic effect between the two metals, which can improve the sulfur resistance of the catalyst to some extent.

Zhang et al. [40] found that the Co-modified Ce/ MnO_2 catalyst exhibited excellent SO_2 tolerance due to the addition of Co, which inhibited the sulfation of CeO_2 and MnO_2 to CeSO_4 and MnSO_4 relieved the reduction of chemisorbed oxygen, and restrained the chemisorption of SO_2 on the catalyst.

Hao et al. [41] prepared a Sn-doped CeMoO_x catalyst that revealed better redox ability and higher Brønsted acid sites, which greatly reduced the adsorption of SO_2 .

MnSO_4 formation on the $(\text{Cu}_{1.0}\text{Mn}_{2.0})_{1-\delta}\text{O}_4$ spinel was significantly alleviated by Cu doping, mainly due to the reduction in the amount of adjacent Mn. Furthermore, because of electron transfer between Cu and Mn cations within the spinel lattice ($\text{Cu}^{2+} + \text{Mn}^{3+} \rightleftharpoons \text{Mn}^{4+} + \text{Cu}^+$), the $(\text{Cu}_{1.0}\text{Mn}_{2.0})_{1-\delta}\text{O}_4$ spinel maintained a high ratio of $\text{Mn}^{4+}/\text{Mn}_{\text{total}}$ on the surface and good low-temperature SCR activity under SO_2 -containing conditions [42].

Jiang et al. [43] doped V_2O_5 into the Mn-Ce(0.4)/AC catalyst to enhance the surface acidity of the catalyst and enrich its surface chemisorbed oxygen, which accelerated the SCR reaction. The accelerated SCR reaction and stronger surface acidity inhibited the competitive adsorption of SO_2 and limited the reaction between adsorbed SO_2 and NH_3 species. Furthermore, vanadium oxide clusters over the catalyst partially prevented sulfation of the dispersed Mn-Ce solid solution by SO_2 .

The MnCrO_x /Sepiolite catalyst has more acidic sites and oxygen vacancies, as well as a stronger redox ability compared to the pure MnO_2 /Sepiolite catalyst. It was found that the addition of Cr could reduce the adsorption strength of SO_2 on the catalyst surface, thus increasing the resistance of the catalyst to SO_2 poisoning and reducing the activation energy [44].

In addition, Wu et al. [45,46] revealed that catalysts with a Keggin structure possessed more surface Brønsted and Lewis acid sites. These catalysts showed significantly improved performance in SCR reaction and resistance to SO_2 poisoning, mainly because the super acidity and Keggin structure inhibited SO_2 adsorption.

Wang et al. [47] found that the addition of Fe and Co to Mn-Ce/TiO₂ significantly reduced the adsorption of SO_2 . The uniform distribution of Fe and Co on the surface of Mn-Ce/TiO₂ prevented the diffusion of SO_2 to the inner layer of the catalyst. In the presence of SO_2 , the SCR reaction over 2Fe₄Co-MCT catalyst mainly followed the Eley–Rideal (E–R) mechanism, which the adsorbed NH_3 and NH_4^+ species reacted with gaseous NO species to produce unstable NH_4NO_2 , followed by the rapid decomposition to N_2 and H_2O .

Inhibiting the oxidation of SO_2 to SO_3 can further avoid the formation of NH_4HSO_4 , resulting in a reduction in the SO_2 poisoning over the catalyst.

Wang et al. [48] reported that doping Sm into the MnCeTiO_x catalyst could increase oxygen vacancies and transfer electrons to Mn^{4+} and Ce^{4+} , which facilitates the formation of active adsorbed NO_2 , binary nitrate, and bridging nitrate intermediates. Meanwhile, it inhibited the oxidation of SO_2 by Mn^{4+} and Ce^{4+} , leading to the suppression of SO_2 poisoning of the catalyst (Figure 1).

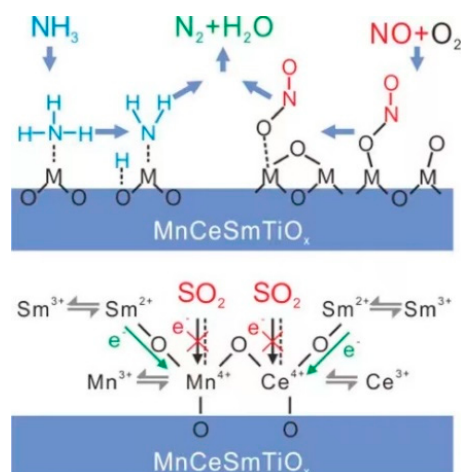


Figure 1. Proposed mechanism for the SCR reaction on MnCeTiO_x. Proposed mechanism for the suppressed oxidation of SO_2 to SO_3 on the Sm-modified MnCeTiO_x. These schematic diagrams are reproduced with permission from [48]. Copyright: 2020 American Chemical Society.

Hou et al. [49] prepared a La-modified TiO₂ carrier by the sol–gel method, and then prepared a Fe-Mn/TiO₂(xLa) catalyst, which significantly improved SO_2 resistance. It is mainly accounted for the fact that the addition of La inhibited the growth of TiO₂ particles, which resulted in a distinct layered structure and more mesopores of TiO₂, thus accelerating the decomposition of ammonium sulfate. Meanwhile, the increase in Brønsted acid sites and the electron transfer between La and Fe/Mn in the La-doped catalyst inhibited the adsorption and oxidation of SO_2 on the catalyst, weakened the sulfation of the active components, and thus improved SO_2 resistance.

Zhu et al. [50] prepared a Ce-based catalyst by the solid-state ball milling method using $\text{Ce}_2(\text{C}_2\text{O}_4)_3$ as a precursor. It was found to have a large specific surface area and pore volume. In addition, there was a large amount of Ce^{3+} and surface-adsorbed oxygen, which promoted the adsorption and activation of NO. Stronger Lewis acid sites resulted in the enhanced adsorption of NH_3 and the reduced adsorption and oxidation of SO_2 .

Liu et al. [51] found that, by embedding Mn₃O₄ nanoparticles in a defect-rich graphitic carbon framework (Mn₃O₄@G-A) through a MOF-derived confinement–decomposition–oxidation strategy, electron transfer from graphitic carbon to Mn₃O₄ through Mn–O–CGD bonds modulated the electronic properties of Mn₃O₄, resulting in a higher vacancy for-

mation energy and weaker oxidation ability of $\text{Mn}_3\text{O}_4@\text{G-A}$, which in turn improved SO_2 resistance.

Wu et al. [8] used a high shear method to uniformly mix TiO_2 particles with EG as a carrier and then loaded vanadium, which increased the surface area and the number of Brønsted acid sites over the catalyst and improved the surface VO^{2+} as well as adsorbed oxygen species, thus reducing the adsorption and oxidation of SO_2 .

4.1.2. Increasing the Adsorption of Active Intermediate Species with the Coexistence of Sulfate Species

Xu et al. [52] showed that K-modified activated carbon enhanced the adsorption of NO. Furthermore, the additional formation of nitro compounds, singlet nitrate and nitrous oxide promoted the denitrification reaction pathway. The catalytic dissociation activity of N_2O on AC-K brought the formation and decomposition of N_2O into a dynamic equilibrium. Thus, it provided a good resistance to sulfur dioxide.

Regarding the Ho-modified Mn/Ti catalyst, the influence of SO_2 on the adsorption and activation of NO on the catalyst surface was weak, and the adsorption and activation of NH_3 were not affected, only the reactions obeying the L-H mechanism were inhibited. Therefore, the 0.2HoMn/Ti catalyst had a better SO_2 resistance [53].

Yu et al. [54] found that NH_4^+ in NH_4HSO_4 deposited on the surface of the $\gamma\text{-Fe}_2\text{O}_3$ catalyst readily reacted with $\text{NO} + \text{O}_2$, which suppressed the continued accumulation of NH_4HSO_4 , and an equilibrium between the deposition and consumption of NH_4HSO_4 on the surface of $\gamma\text{-Fe}_2\text{O}_3$ was established. While SO_4^{2-} from ammonium sulfate continued to combine with iron ions to form iron sulfate on the catalyst surface, the formation of iron sulfate species increased the number of acid sites and enhanced the acidity intensity, thus facilitating the E-R reaction pathway. The amount of oxygen species adsorbed on the surface also increased. All of these processes are important in enhancing the efficiency of SCR.

4.1.3. Building Sacrificial Sites to Conserve Active Sites

The reaction of SO_2 and active CeO_2 to form $\text{Ce}_2(\text{SO}_4)_3$ is the decisive reason for SO_2 poisoning on the Ce-based catalyst. The addition of Fe_2O_3 to the Ce- Nb_2O_5 catalyst made it possible to act as a sacrificial victim, which reacted preferentially with SO_2 , thus releasing the active species [55]. Kang et al. [56] also found, in this phenomenon, that the addition of Fe_2O_3 , in the presence of SO_2 , protected the active CeO_2 , inhibited sulfate deposition and improved the catalytic performance of the wollastonite-supported $\text{CeO}_2\text{-WO}_3$. Furthermore, the addition of Fe can increase the surface acidity, surface oxygen mobility and the amount of surface adsorbed oxygen of the catalyst. Meanwhile, the SO_4^{2-} formation rate can be greatly reduced, thus reducing the sulfur poisoning of the catalyst [57].

In addition, Cao et al. [58] revealed that loading niobium on CeWTi complex oxides reduces the formation of metal sulfates and protects Ce^{x+} from sulfur dioxide attacks. Wang et al. [59] discovered that CeO_2 -modified MnCoO_x microflowers (MnCoCeO_x) with increased ratios of $\text{Ce}^{3+}/\text{Ce}^{n+}$, $\text{Mn}^{4+}/\text{Mn}^{n+}$ and $\text{O}_\alpha/(\text{O}_\alpha + \text{O}_\beta)$ prevented the sulfation of MnCoO_x into metal sulfate species and improved the anti-sulfur properties of the catalyst.

Fabricated nanomaterials with a core-shell structure or a special structure can usually serve as a protective layer for the active component. Cai et al. [60] found that the Fe_2O_3 shell significantly inhibited the deposition of sulfate species on the surface of multi-shell $\text{Fe}_2\text{O}_3@\text{MnO}_x@\text{CNTs}$ as compared with $\text{MnO}_x@\text{CNT}$ catalyst, showing excellent SO_2 tolerance. Shao et al. [61] showed that H- MnO_2 with a hollow cavity structure could provide shuttle space for oxidation and improved mass adsorption and conversion, which can lead to better NO conversion and SO_2 resistance. Table 1 lists some typical SO_2 -tolerant SCR catalysts in the literatures are summarized as following.

Table 1. Typical SO₂-tolerant catalysts reported in the literature.

Catalysts	Reaction Condition	NO Conversion before and after Introducing SO ₂	Reference
Sn/0.2-CeMoO _x	T = 225 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 5%, [SO ₂] = 200 ppm, GHSV = 100,000 h ⁻¹	Decline from 99% to 91%	[41]
(Cu _{1.0} Mn _{2.0}) _{1-δ} O ₄	T = 200 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 3%, [SO ₂] = 50 ppm, [H ₂ O] = 5 vol%, GHSV = 100,000 h ⁻¹	Maintained at ~87%	[42]
Mn-Ce(0.4)-V/AC	T = 200 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 5%, [SO ₂] = 100 ppm, [H ₂ O] = 10 vol%, GHSV = 18,000 h ⁻¹	Maintained at ~80%	[43]
MnCrO _x /Sepiolite	T = 220 °C, [NH ₃] = [NO] = 1000 ppm, [O ₂] = 5%, [SO ₂] = 200 ppm, GHSV = 35,000 h ⁻¹	Decline from 87% to 85%	[44]
V ₂ O ₅ -MoO ₃ /TiO ₂	T = 250 °C, [NH ₃] = [NO] = 1000 ppm, [O ₂] = 5%, [SO ₂] = 1000 ppm, [H ₂ O] = 10 vol%, GHSV = 40,000 h ⁻¹	Decline from 100% to 83%	[45]
2Fe ₄ Co-MCT	T = 200 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 6%, [SO ₂] = 200 ppm, [H ₂ O] = 10 vol%, GHSV = 12,000 h ⁻¹	Decline from 98% to 90%	[47]
MnCeSmTiO _x	T = 200 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 5%, [SO ₂] = 200 ppm, [H ₂ O] = 5 vol%, GHSV = 80,000 h ⁻¹	Maintained at ~70%	[48]
Fe-Mn/TiO ₂ (0.02La)	T = 200 °C, [NH ₃] = [NO] = 1000 ppm, [O ₂] = 7%, [SO ₂] = 100 ppm, GHSV = 30,000 h ⁻¹	Maintained at ~99%	[49]
CeTiO _x	T = 300 °C, [NH ₃] = [NO] = 1000 ppm, [O ₂] = 6%, [SO ₂] = 175 ppm, [H ₂ O] = 6 vol%, GHSV = 30,000 h ⁻¹	Decline from 100% to 70%	[50]
Mn ₃ O ₄ @G-A	T = 160 °C, [NH ₃] = 660 ppm, [NO] = 600 ppm, [O ₂] = 5%, [SO ₂] = 50 ppm, GHSV = 20,000 h ⁻¹	Decline from 100% to 70%	[51]
0.2HoMn/Ti	T = 180 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 6%, [SO ₂] = 100 ppm, [H ₂ O] = 10 vol%, GHSV = 20,000 h ⁻¹	Decline from 100% to 80%	[53]
CeO ₂ -Fe ₂ O ₃ -Nb ₂ O ₅	T = 300 °C, [NH ₃] = [NO] = 667 ppm, [O ₂] = 5%, [SO ₂] = 200 ppm, GHSV = 120,000 mL·g ⁻¹ ·h ⁻¹	Maintained at ~92%	[55]
Fe(4)@CeW/H	T = 300 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 5%, [SO ₂] = 100 ppm, [H ₂ O] = 8 vol%, GHSV = 40,000 h ⁻¹	Decline from 100% to 82%	[56]
Mn ₁ Fe _{0.25} Al _{0.75} O _x	T = 150 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 5%, [SO ₂] = 50 ppm, GHSV = 60,000 h ⁻¹	Maintained at ~80%	[57]
NbCeWTi	T = 270 °C, [NH ₃] = [NO] = 500 ppm, [O ₂] = 5%, [SO ₂] = 200 ppm, [H ₂ O] = 5 vol%, GHSV = 80,000 h ⁻¹	Decline from 92% to 71%	[58]
Fe@Mn@CNTs	T = 240 °C, [NH ₃] = [NO] = 550 ppm, [O ₂] = 5%, [SO ₂] = 100 ppm, [H ₂ O] = 10 vol%, GHSV = 20,000 h ⁻¹	Decline from 100% to 91%	[60]

4.1.4. Promoting the Decomposition of Sulfates

Some additives can facilitate the decomposition of surface (NH₄)₂SO₄ and NH₄HSO₄, leading to the improvement of the sulfur resistance of the catalyst. Fan et al. [62] found that Al₂O₃ added into MnO_x not only inhibited the reaction between MnO_x and SO₂ and promoted the decomposition of NH₄HSO₄, but also led, to a certain degree, to a decrease in the thermal stability of the adsorbed SO₂ species. The CeO₂ or WO₃ incorporated into a V₂O₅/TiO₂ catalyst could promote the decomposition of the NH₄HSO₄ deposited on the catalyst surface [63,64]. Additionally, Ye et al. [65] revealed that Nb₂O₅ and Sb₂O₅ doped into the V₂O₅-WO₃/TiO₂ catalyst could accelerate the reaction of the deposited NH₄HSO₄ with gaseous NO over the catalyst.

Furthermore, Guo et al. [66] also found that the larger pore size of the carrier could greatly ease the deposition of ABS. Using mesoporous silica SBA-15 as the support material, the decomposition of ABS was significantly accelerated with the enlarged pore size, largely because the larger pore size generated higher vapor pressure, which facilitated the vaporization and decomposition of ABS (Figure 2).

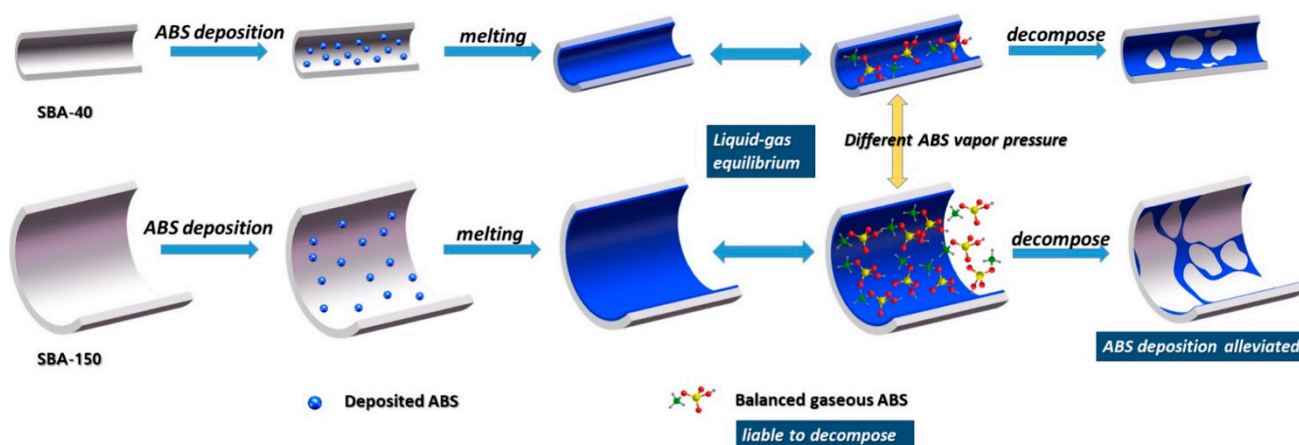


Figure 2. Illustration of ABS decomposition behaviors on SBA-15 with different pore sizes. These schematic diagrams are reproduced with permission from [66]. Copyright: 2019 American Chemical Society.

4.2. H_2O Poisoning

The effect of H_2O on SCR activity is mainly due to the following three reasons. (i) The competitive adsorption of H_2O and NO or NH_3 decreased the NH_3 -SCR reaction activity. (ii) H_2O could promote the formation of hydroxyl groups (OH) on the catalyst surface, thus reducing the number of active sites and inhibiting the adsorption of NH_3 and NO species and the reaction between them [67]. (iii) H_2O could accelerate the formation of NH_4HSO_4 when both SO_2 and H_2O are present simultaneously. There are several ways to reduce the adsorption of H_2O on the SCR catalyst surface and the active center to improve the H_2O resistance of the catalyst [68].

4.2.1. Doping with Rare Earth Metal Elements or Transition Metal Elements

Zhang et al. [69] gave an example to improve the H_2O tolerance in Mn-based materials. The Ce-MnO₂ catalyst has good resistance to H_2O , and both Ce-containing catalyst has a more than 90% NO_x conversion below 150 °C in the presence of superior H_2O . The competitive adsorption of NO_x on the surface of Ce-MnO₂ was stronger than that of H_2O . However, NH_3 and H_2O did not have any competitive adsorption on both. Ma et al. [70] reported that doping Ce on the Cu_{0.02}Fe_{0.2}TiO_x catalyst increased the H_2O resistance of the catalyst dramatically, mainly because the amount of Lewis acid increased largely and the interaction between H_2O and Lewis acid sites was weaker than that of Brønsted acid sites.

Guo et al. [71] added Fe to the Mn-Eu catalyst, which showed excellent resistance to H_2O , even at a high gas hourly space velocity (GHSV) of 75,000 h^{−1}. The NO conversion remained at 90% at 230 °C in the presence of 15% H_2O after 50 h, and the detrimental effect of H_2O could be quickly eliminated after its removal.

4.2.2. Doping or Modification of the Catalyst Surface with Hydrophobic Materials or Moieties

Zhang et al. [72] showed a way to enhance the H_2O resistance with the assistance of hydrophobic polytetrafluoroethylene (PTFE) materials. The Mn-PTFE composite catalyst exhibited a good tolerance to H_2O , which was related to the hydrophobic surface generated, making the catalyst surface active sites less likely to be consumed by H_2O . Additionally, Wang et al. [73] found that polyethylene oxide-polypropylene-polyethylene oxide (P123)-modified Mn-MOF-74 demonstrated excellent NO conversion of up to 92.1% in the presence of 5% H_2O at 250 °C. The undecorated Mn-MOF-74, under the same conditions, obtained only 52% NO conversion. Alternatively, the introduction of a hydrophobic group ($-CH_3$) on the ligand of Mn-MOF-74 could similarly reduce the effect of H_2O .

4.2.3. Regulating the Crystal Phase and Structure

Hu et al. [74] found that α -MnO₂ has a better resistance to H₂O than β -MnO₂. Because α -MnO₂ has a special semi-tunnel structure and exposed active (110) surface, a double ionic cluster can be easily formed over the surface of α -MnO₂, showing good hydrophobicity in the presence of H₂O. Qiu et al. [75] showed that 3D-MnCo₂O₄ has a good resistance to H₂O and about 96% NO conversion can be achieved over the catalyst at 200 °C in the presence of 10% H₂O. Furthermore, Tang et al. [76] revealed that the resistance of H₂O over MnO_x was also related to the synthesis method. MnO_x prepared by the Co-precipitation method has a better H₂O resistance than MnO_x prepared by the solid phase reaction method at 80 °C (10% H₂O, 47,000 h⁻¹) because the former has a larger specific surface area.

4.3. Alkali Poisoning

An alkali metal has a toxic effect on the catalyst mainly because it occupied the acidic sites of the catalyst, reducing the number of acidic sites and decreasing the adsorption of NH₃ [77,78]. The following two approaches are available to reduce the toxicity of alkali metals to catalysts.

4.3.1. Providing More Acidic Sites

The deposition of an acidic promoter on the catalyst surface can increase the number of acidic sites on the surface as well as binding to toxic metals, leaving the active metal sites free, thus improving the resistance of SCR catalysts to alkali metals [79]. Kang et al. [80] indicated that CuO has an excellent reducing property and surface acidity and the CuO modification on CeTiO_x catalyst effectively improved its low-temperature catalytic activity and resistance to alkali metal poisoning. Co modification on 0.3K3Mn10Fe/Ni catalyst resulted in the increase in the ratio of NiO/Ni, the concentration of Fe³⁺ and Mn⁴⁺, lattice oxygen percentage, unsaturated Ni atoms content, and the number of Ni-Mn/Fe composite oxides structure defects, which caused an enhancement of the synergistic effect between Ni and Mn/Fe. The increase in Brønsted acid sites results in the enhancement of the mid-low temperature redox ability of the catalyst, thus significantly improving the K poisoning resistance of the 3Mn10Fe/Ni catalyst. For the 3Mn10Fe/Ni catalyst, as the addition of Co is 0.2%, the K poisoning resistance of the catalyst is optimal, and the de-NO_x activity reaches 98% at 220 °C [81]. Nie et al. [82] discovered that V₂O₅-Ce(SO₄)₂/TiO₂ performs better in a SCR reaction than V₂O₅-WO₃/TiO₂ when KCl is deposited on the catalyst surface. This is because the V₂O₅-Ce(SO₄)₂/TiO₂ catalyst has a stronger surface acidity. Moreover, Wang et al. [83] clearly identified that the acidic H-SAPO-34 with abundant Brønsted acid sites ensured a high dispersion of active Cu²⁺ sites, while providing a large number of ion-exchange protons to replace the K, Ca, Pb ions in the zeolite framework. The Ce species promoted the electron-transfer effect with Cu active sites and improved redox cycling. It could also bind to Ca and Pb toxicants and promote the resistance of the catalysts to harmful elements (Figure 3). Accordingly, the dual promotion of H-SAPO-34 and Ce enabled the CuCe/H-SAPO-34 catalyst to effectively control NO_x emissions in the presence of alkali metals.

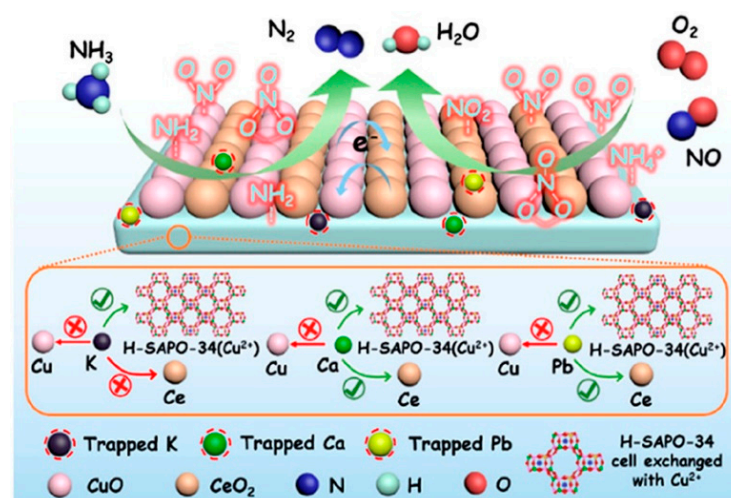


Figure 3. Proposed mechanism of NO_x reduction in the presence of alkaline and heavy metals over CuCe/H-SAPO-34 catalysts. These schematic diagrams are reproduced with permission from [83]. Copyright: 2020 American Chemical Society.

4.3.2. Building Alkaline Capture Sites to Protect Active Sites

Zha et al. [84] reported that the Fe-OMS-2 catalyst was found to have a better alkaline tolerance due to having more redox species, more acidic sites and a stronger adsorption of NO_x species as compared with the undoped MnO_x octahedral molecular sieve (OMS-2) catalyst. Meanwhile, K⁺ ions can be trapped by cryptic tunneling through an ion exchange mechanism, resulting in a more stable system structure. Wu et al. [85] synthesized an alkali metal-resistant deNO_x catalyst that utilized ion-exchanged titanate nanotubes as supports to stabilize alkali metal ions in the interlayer of titanate nanotubes. Yan et al. [86] proved that the OMS-5(H)@TiO₂ catalyst has a good resistance to alkali metals. The special structure of the MnO_x octahedral molecular sieve (OMS-5(H)) could trap alkali metals and the acid treatment increased the acid sites, which enabled the catalyst to resist alkali metals in the form of ion exchange. The decoration of TiO₂ further increased the strength of Lewis acid sites, allowing more active intermediate species to participate effectively in the NO_x reaction.

The CuNbTi complex oxide catalyst exhibited a good alkali resistance. The K atom preferentially interacted with Nb–OH and Nb=O, which protected the active species. K₂O can be trapped on TiO₂ supported by the addition of Niobium species, while isolating Cu²⁺, as the active sites play a key role in yielding the remaining activity [87].

The works discussed above have laid the foundation for the development of superior low-temperature SCR catalysts, and much progress has been made in catalyst resistance to poisoning. However, successful applications in flue gas containing one or more components of SO₂, H₂O, and alkali metals have rarely been reported. The tolerance of SO₂ and H₂O is closely related to the reaction temperature, and most experiments have been performed above 200 °C, which may not have practical applications. At least 10% or even 30% of H₂O is actually present in flue gas. The experimental H₂O content and stability testing time are often insufficient. Researchers have studied the effect of alkali on SCR catalysts mainly by impregnating the catalyst with alkali metal precursors followed by a calcination treatment. The deactivation process of alkali in fly ash in actual flue gas may be different from the alkali poisoning process of the impregnation method. Therefore, bringing the anti-poisoning catalysts closer to practical applications is a challenge and more efforts are needed in this area.

5. Industrial Applications of Low-Temperature SCR Catalysts in China

In the last decade, the research and development of low-temperature SCR catalysts and deNO_x engineering technology have achieved great progress and success in China. Commercial low-temperature SCR catalyst products are widely used in deNO_x project in

the coking, sintering, waste incineration and lime kiln industries. Table 2 lists the low-temperature SCR catalysts produced by major domestic enterprises and their engineering applications, including demonstration projects.

Table 2. Low-temperature SCR catalyst research units and commissioning (test) information [88].

Catalyst	Company	Cooperative Unit	Temperature Window in Which It Is Run	Application Examples
“Fangxin” Vanadium-Titanium Catalyst	Advanced E-catal. Co., Ltd.	Beijing University of Technology	160–400 °C	Zhanjiang Baosteel Group Co., Ltd. 7 M top mounted coking furnace-DeNO _x
Low-temperature honeycomb and foam type non-vanadium catalysts	Zhongneng Guoxin (Beijing) Technology Development Co.	Tsinghua University	150–400 °C	Cement kiln low-temperature SCR denitrification
Mn/FA-PG non-vanadium catalyst	Hefei Chenxi Environmental Protection Engineering Co.	Hefei University of Technology	180–300 °C	Shandong Tiexiong Xinsha Energy Co. Coke oven flue gas
MnO _x -CoO _x (CeO _x)TiO ₂ Honeycomb/stacked bar catalysts	Shanghai Han Yu Environmental Materials Co.	Zhejiang University	130–260 °C	Glass kiln and petrochemical field

The first successful application of low-temperature SCR is the 7M top mounted with 4×65 hole coking furnace deNO_x project located in Zhanjiang Baosteel Group Co., Ltd., which also is the world’s first industrialized deNO_x project using a low-temperature SCR catalyst with a running temperature of 180–200 °C for the purification of the coking flue gas [89].

The project was designed to produce about 3.4 million tons of dry coke per year at the largest scale, using 4×65 -hole JNX2-70-2 type reheat top-loading coke ovens. It required to simultaneously control SO₂, NO_x and PM emissions in flue gas to meet strict emission regulations. As shown in Figure 4, the flue gas flows through the SDA (Spray Drying Absorption) desulfurization system and the integrated reactor of the dust filter close-coupled with the SCR catalyst, and then finally returns to the chimney. The SCR reactor is designed with 16 independent reaction chambers, each of which can be alternately individually heated to 350 °C to decompose the NH₄HSO₄ generated in the reaction, realizing the in situ thermal regeneration of the SCR catalyst and ensuring the normal operation of the low-temperature SCR catalyst.

The engineering purification equipment of this project was formally put into operation in November 2015, and to date, the flue gas SO₂ emission concentration decreased from 80–150 mg/Nm³ to 20 mg/Nm³, the total SO₂ emission decreased from 180 kg/h to 24 kg/h, the desulfurization efficiency reached 90%, cutting SO₂ by about 30,000 tons/year. The NO_x emission concentration decreased from ca. 500 mg/Nm³ to ca. 100 mg/Nm³, the total NO_x emission decreased from 600 kg/h to 120 kg/h with deNO_x efficiency of ca. 80 %, and the NO_x emission was reduced by about 100,000 tons/year, resulting in significant environmental, social and economic benefits. The catalyst used in this project is a V–Ti-based low-temperature SCR catalyst developed by He’s group at Beijing University of Technology. This catalyst was officially produced and put into the market in 2012 by Advanced E-catal. Co. Ltd., located in Beijing, China. In addition to the application of coking flue gas denitration, the low-temperature SCR catalyst was also used in the purification of sintering flue gas. Due to the large volume of flue gas, complex pollutants, large working condition fluctuates, low flue gas temperature and high sulfur content, the denitrification of sintering flue gas faces huge technical challenges. Renfon Steel Co., Ltd., located in Tangshan, employed the engineering process of “semi-dry desulfurization of flue

gas + GGH (Gas Gas Heater) heat exchange + flue gas warming + low temperature SCR denitrification + GGH heat exchange + induced draft fan + stack emission” to control the emission of sintering flue gas. The project with low-temperature SCR denitrification was successfully put into operation to meet the strict emission regulations on 2 November 2018, using the “Fangxin” low-temperature catalyst produced by Advanced E-catal. Co. Ltd. This is the first application of a low-temperature SCR denitrification catalyst on a sintering flue gas denitrification project in China [90]. The inlet NO_x concentration of the SCR reactor is ca. $200\text{--}300\text{ mg/Nm}^3$ and the outlet NO_x concentration is stabilized below 20 mg/Nm^3 with ammonia escape smaller than 1 ppm, which can realize the ultra-low emission of sinter flue gas NO_x or even the near-zero emission requirement in the future. The project also showed that the low-temperature SCR catalyst can eliminate the dioxin emissions in its application. The project was a huge success as all pollutant emissions are lower than the values stipulated by the national environmental regulations, meeting the expected targets.

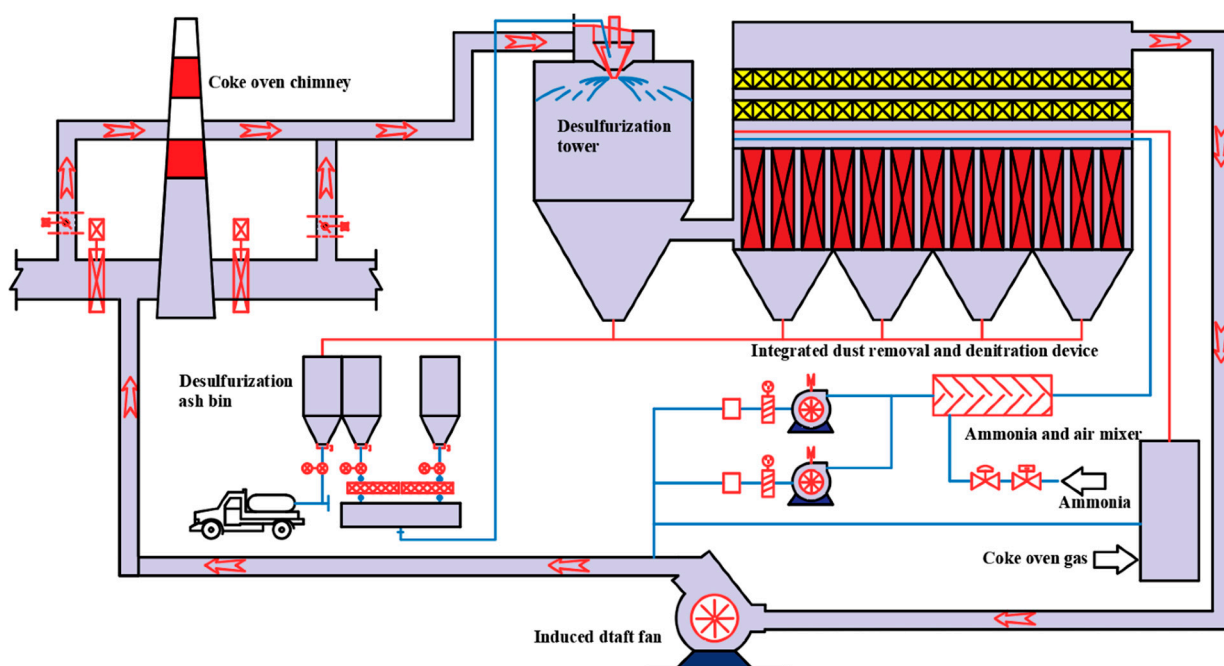


Figure 4. Coking de NO_x project flow chart using the low-temperature SCR catalyst in Zhanjiang Baosteel Group Co.

6. Concluding Remarks and Future Perspectives

Low-temperature SCR technology has been proven as an effective and crucial way to abate the NO_x emitted from non-electric industries, based on its several successful applications in China in the past years. There are still many problems and challenges in gaseous pollutant control in non-power industries, which need to be studied further.

Moreover, new technology with a lower energy consumption and multi-functions needs to be developed to update the demands of deep treatments of NO_x . The following issues need to be addressed in future research:

- (1) The SO_2 poisoning of the low-temperature SCR catalyst is still a vast problem in practice, especially in flue gas with high humidity.
- (2) NH_3 slips from the de NO_x system, releasing ammonium salts to the environment. The selective oxidation of the NH_3 catalyst should be developed, which should be coupled with the SCR catalyst in practical applications.
- (3) In some industries, there may also be harmful elements, such as As, Hg, and Pd, in flue gas, which will affect the service life of the SCR catalysts, but also cause air pollution. Therefore, in the design of the catalyst and pollution control engineering,

new technologies should be developed to solve the complex smoke pollution problems with, for example, the development of multi-functional SCR catalysts.

- (4) In flue gas, such as sintering, in addition to NO_x , there is a relatively high concentration of CO. How to use the oxidation of CO to increase the temperature of the SCR catalyst bed or how to use CO as a reducing agent to treat NO_x are also very important topics that need to be studied in the future.

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