

Review

A Review of Synergistic Catalytic Removal of Nitrogen Oxides and Chlorobenzene from Waste Incinerators

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Abstract: Emission of harmful gases, nitrogen oxides (NO_x), and dioxins pose a serious threat to the human environment; so, it is urgent to control NO_x and dioxin emissions. The new regulations for municipal solid waste incineration emissions set new stringent requirements for NO_x and dioxin emission standards. Most of the existing pollutant control technologies focus on single-unit NO_x reduction or dioxin degradation. However, the installation of separate NO_x and dioxins removal units is space-consuming and costs a lot. Nowadays, the simultaneous elimination of NO_x and dioxins in the same facility has been regarded as a promising technology. Due to the extremely high toxicity of dioxins, the less toxic chlorobenzene, which has the basic structure of dioxins, has been commonly used as a model molecule for dioxins in the laboratory. In this review, the catalysts used for nitrogen oxides/chlorobenzene (NO_x/CB) co-removal were classified into two types: firstly, non-loaded and loaded transition metal catalysts, and their catalytic properties were summarized and outlined. Then, the interaction of the NH₃-SCR reaction and chlorobenzene catalytic oxidation (CBCO) on the catalyst surface was discussed in detail. Finally, the causes of catalyst deactivation were analyzed and summarized. Hopefully, this review may provide a reference for the design and commercial application of NO_x/CB synergistic removal catalysts.

Keywords: nitrogen oxides; dioxins; chlorobenzenes; synergistic removal; interaction



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1. Introduction

In the process of urbanization, the accumulation of solid waste has posed a serious threat to the ecological environment. The methods to deal with solid waste mainly include sanitary landfill [1], waste incineration [2], and high-temperature composting [3]. Among them, waste incineration technology has become the mainstream direction of municipal solid waste treatment due to its advantages such as obvious volume reduction of solid waste, strong destruction of organic toxicity, and recycling of incineration heat energy [4,5]. However, waste incineration exhaust gas contains secondary pollutants such as nitrogen oxides (NO_x), sulfur dioxide, carbon monoxide, hydrogen chloride, heavy metals, and dioxins [6,7], and their hazards should not be underestimated. In particular, NO_x and dioxin emissions have caused widespread concern. NO_x (mainly including NO and NO₂) is one of the main causes of acid rain, photochemical smog, ozone depletion, and eutrophication [8–10]. Dioxins are a generic term for polychlorinated biphenyldioxins (PCDDs) and polychlorinated biphenyldioxins and furans (PCDFs). Dioxins are extremely toxic, chemically stable, metabolize extremely slowly after entering the human body, and have been recognized as carcinogens [11,12]. In order to protect the environment and safeguard human health, countries have put forward higher requirements for NO_x and dioxin emission standards in the new regulations on exhaust gas emissions. Among them, China has revised the emission standards for tail gas pollutants from waste incineration plants,

tightening the dioxin emissions from 1.0 ngTEQ/Nm³ to 0.1 ngTEQ/Nm³ and strictly specifying the control standards for NO_x [13].

The main methods to control NO_x emissions include adsorption [14], ionophore activation [15], selective catalytic reduction (SCR) [16], and selective non-catalytic reduction (SNCR) [17,18]. Among them, NO_x removal by catalytic materials using NH₃ as a reducing agent (NH₃-SCR) is the most efficient and environmentally friendly and has been widely used industrially [19]. In practical applications, NH₃-SCR catalysts are classified into three types: low-temperature catalysts, medium-temperature catalysts, and high-temperature catalysts. Their detailed use temperatures and use characteristics are shown in Table 1. Current studies have found that most catalysts followed Eley–Rideal (E-R) and Langmuir–Hinshelwood (L-H) reaction mechanisms in the NH₃-SCR reaction [20,21]. The E-R mechanism refers to the reaction between the gas phase NO_x and the adsorbed state NH₃; the L-H mechanism refers to the reaction between the adsorbed state NO_x and the adsorbed state NH₃ [22]. The main chemical reactions of the NH₃-SCR process can be summarized as follows [23,24]:

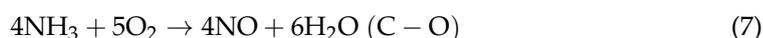
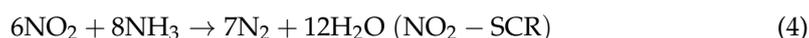
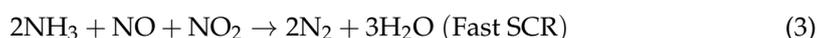
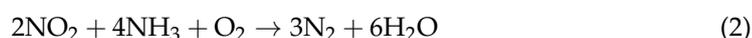


Table 1. Activity characteristics of three types of industrial NH₃-SCR catalysts.

Catalyst Type	Operating Temperature (°C)	Main Features
High-Temperature Catalyst	345–595	High NO _x conversion rate; less leakage of NH ₃ ; strong resistance to SO ₂ poisoning above 425 °C
Medium-temperature catalyst	260–425	Wide application and high denitrification efficiency
Low-temperature catalyst	150–300	Low operating temperature, low energy consumption

Among them, (1) and (2) are the main reactions in NH₃-SCR. The “fast SCR” is more efficient than the “standard SCR” at 200 °C [25]. The process of NO oxidation to NO₂ is the rate-limiting step of the “fast SCR” [24,26]. The rate of the “NO₂-SCR” is much lower than that of the “standard SCR”. The production of N₂O in the “NSCR” is the main reason for the reduced selectivity of N₂. In the SCR process, the strong oxidation of the catalyst at high temperature leads to a direct redox reaction between NH₃ and O₂, resulting in the inability of NH₃ to react with NO, a reaction process defined as the C-O reaction. In the C-O reaction (7), NH₃ is inactivated by the over-oxidation of O₂. This is the main reason for the decrease in NO_x conversion rates in a high-temperature environment [27]. The effect of the NO to NO₂ ratio and temperature on the NH₃-SCR process is shown in Figure 1 [24]. When designing catalysts for different temperature conditions, care should be taken to bias the reaction toward fast SCR while avoiding NO₂-SCR, NSCR, and C-O.

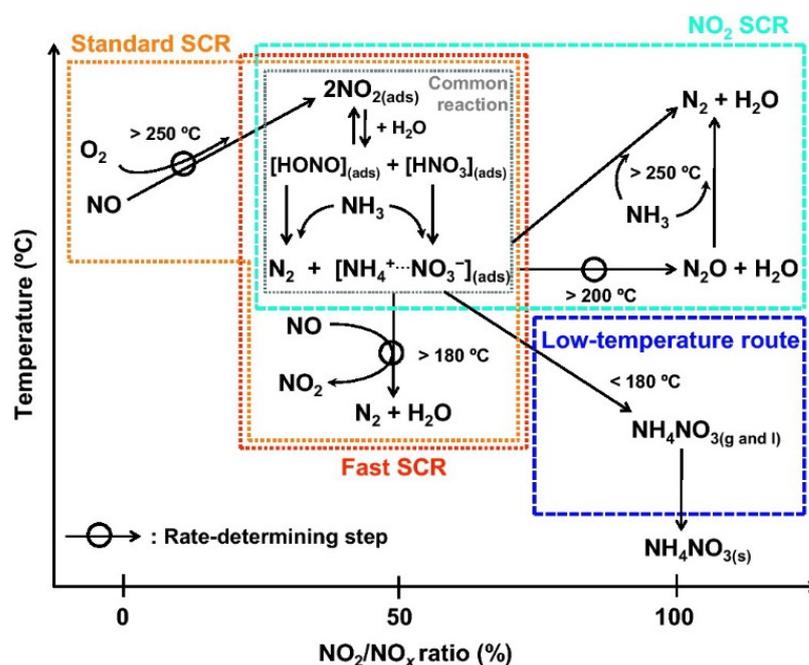


Figure 1. Effect of NO to NO₂ ratio and temperature on NH₃-SCR reaction [24]. Copyright 2010, Elsevier B.V.

The degradation pathways of dioxins mainly include microbial degradation [28,29], photodegradation [30,31], and thermocatalytic degradation [32–34]. Thermocatalytic degradation is the conversion of dioxins into harmless products (CO₂, CO, and H₂O) and easily removable products (HCl and Cl₂) by using catalysts to accelerate the dechlorination and degradation process of dioxins under heating conditions [33]. This technology has the advantages of lower reaction temperature and higher dioxin removal efficiency and is considered the best dioxin removal pathway. Dioxins are very difficult to study because of their complex structure, huge toxicity, and high price. Chlorobenzene, which has low toxicity, cheap cost, and a structure similar to that of dioxins, is commonly used in experiments to replace dioxins [35]. The first step of CB catalytic oxidation is the adsorption of CB on the catalyst surface. The second step is dechlorination, in which Cl⁻ is attacked by nucleophiles and replaced by O₂⁻. The third step is ring opening, the aromatic ring generated after the dechlorination of chlorobenzene is oxidized by electrophilic substitution [36]. The degradation mechanism of CB over Mn_{0.8}Ce_{0.2}O₂/H-ZSM5 catalyst is shown in Figure 2 [37]. CB is adsorbed on the catalyst surface. Then, the C-Cl bond and Brønsted acidic site undergo nucleophilic substitution reaction with the neighboring oxides or hydroxyl radicals, during which the C-Cl bond breaks to generate phenol salts and, in turn, intermediates such as benzoquinone and cyclohexanone are produced. These intermediates are converted by O₂ nucleophilic attack to maleic acid and aldehydes and are eventually deeply oxidized to end products such as CO₂, H₂O, and HCl. In the catalytic oxidation of chlorobenzene, the redox properties, surface acidity (Brønsted and Lewis acid sites), surface adsorbed oxygen, and mobility of lattice oxygen play important roles in CB catalytic combustion [36,38].

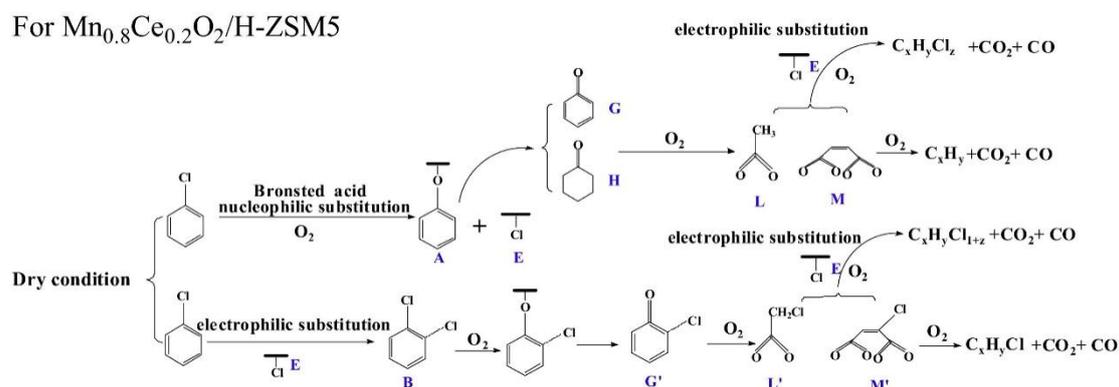


Figure 2. Degradation mechanism of CB literature [37]. Copyright 2016, Elsevier B.V.

Most of the existing pollutant control technologies focus on stand-alone NO_x emission reduction or dioxin degradation. However, setting up single NO_x or dioxin removal units is not only space intensive but also economical and energy intensive. The catalytic conversion of both NO_x and CB utilizes the redox properties and surface acidity of the catalyst, which provides the feasibility of the combined removal of both. The simultaneous removal of NO_x and dioxins in the same facility would have considerably economic and environmental benefits. Therefore, the synergistic removal of NO_x and dioxins has become a hot research topic in recent years. However, the activity of conventional SCR catalysts for the catalytic oxidation of CB (a dioxin substitute) is relatively low, and the selectivity of the products CO , CO_2 , and HCl is even lower than 60%. Therefore, the development of innovative and efficient catalysts is the key to achieving the synergistic removal of NO_x and CB. After masses of scientific research, some results have been achieved in improving the performance of catalysts for efficient synergistic removal. Yin et al. [39] integrated $FeVO_4$ and Fe_2O_3 semiconductor materials into a bifunctional catalyst with stable bifunctional removal of NO_x and CB (>95%) and high HCl selectivity (>85%). Yang et al. [40] prepared Nb-doped $MnCe_{0.2}O_x$ composite oxide catalysts, which achieved 94.5% and 96% removal of NO_x and CB, respectively, at the temperature of 220 °C. However, there are still some scientific issues in the synergistic removal process that have not yet been concluded: (1) the competitive adsorption of multiple reactants on the catalyst active sites and their effects on the catalytic performance; (2) the interaction of multiple gas components in the synergistic removal reaction; (3) the gradual deposition and coverage effects of carbon and halogen species on the catalyst surface; (4) the effects of other components in the waste incineration exhaust gas (SO_2 , heavy metals, H_2O , HCl , etc.) on the catalyst activity. Based on the above, this paper first summarizes the classification of catalysts used for NO_x/CB synergistic removal and analyzes the catalytic properties of each type of catalyst in detail. Then, the mechanism of the interaction between NH_3 -SCR, and CBCO on the catalyst surface is discussed in detail. Finally, the causes of catalyst deactivation are analyzed and summarized.

2. Types of Catalysts for NO and CB Co-Removal

As the core of NO_x and CB synergistic removal technology, catalysts are used to accelerate the reaction, improve the selectivity of N_2 and CO_2 in NO_x and CB end products, and avoid the occurrence of side reactions. The selection of catalysts is crucial. Specifically, qualified NO and CB synergistic removal catalysts should have the following characteristics: (1) high efficiency of NO and CB removal; (2) strong resistance to poisoning; (3) suitable operating temperature range; and (4) high mechanical strength.

Many catalysts have been shown to be active for SCR reactions and CBCO reactions. The common catalysts mainly include noble metal catalysts (Pt, Pb, Ag, etc.), perovskite catalysts (ABO_3 type), and transition metal oxide catalysts (V, Mn, Ce, Co, Fe, Cu, etc.) [38–42]. Although noble metal catalysts have excellent catalytic activity, they cannot be used in large-scale industrial applications because of their high cost, poor stability, weak resistance

to chlorine poisoning, and susceptibility to electrophilic chlorination reactions, which lead to the generation of polychlorine by-products [38,43]. Perovskite catalysts are inexpensive, thermally stable, and have some resistance to poisoning, but they have high activity temperatures and are also susceptible to the generation of polychlorine by-products [41,44]. In contrast, transition metal oxide catalysts are considered ideal for the synergistic catalytic removal of NO_x and CB because of their low cost, high catalytic activity, high selectivity, and strong resistance to poisoning. Herein, the catalytic characteristics of the catalysts are summarized by dividing them into two categories: non-loaded transition metal oxide catalysts and loaded transition metal oxide catalysts.

2.1. Non-Loaded Transition Metal Oxide Catalysts

Transition metal oxides (e.g., CeO₂, MnO_x, CoO_x, VO_x, FeO_x, and CuO_x) are often used as redox centers for the synergistic NO and CB removal reactions due to the valence difference of the metal ions, which leads to good mobility of electrons and lattice oxygen and can significantly reduce the activation energy of the reaction. Further, the metal ions of the catalyst can act as Lewis acid sites, which play an important role in NH₃ and CB adsorption, facilitating the reaction [45]. Moreover, the catalytic performance of single metal oxide catalysts is not satisfactory because of their small specific surface area, poor thermal stability, and their inherent tendency to chlorinate and generate volatile products; therefore, they are less used [46–49]. The addition of dopants to form multi-metal composite oxide catalysts is a common method to improve the defects of single metal oxides. The synergistic removal of NO_x and CB using one catalyst was achieved by rational adjustment of catalyst components. Table 2 shows some non-loaded transition metal oxide catalysts and their synergistic removal efficiencies for NO and CB.

CeO₂ catalysts have received wide attention because of their high oxygen storage capacity and oxygen mobility due to their surface oxygen vacancies and valence transitions between Ce⁴⁺ and Ce³⁺, such as superoxide species (O₂^{•-}) and peroxide species (O₂²⁻), which show good activity in catalytic reactions [50]. However, pure CeO₂ has low resistance to chlorine, poor thermal stability, easy sintering at high temperatures, and reduced oxygen storage capacity, leading to a decrease in catalytic activity [51]. Doping with other transition metals, such as Mn [52], Nb [53], and Zr [54] can effectively increase the rate of Cl removal from the surface of the CeO₂ catalyst while decreasing the energy required for oxygen vacancy generation, improving catalytic activity and stability. Wang et al. [52] showed that the efficiency of MnO_x-CeO₂ composite oxide catalysts for CB removal was much higher than that of CeO₂ and MnO_x single-component catalysts due to the higher mobility of lattice oxygen and surface oxygen of MnO_x-CeO₂, and the large amount of surface active oxygen can effectively remove the Cl components generated at the surface during the catalytic reaction, thus maintaining the high catalytic efficiency of the catalyst. Gan et al. [55] prepared a series of MnO_x-CeO₂ catalysts with different ratios of metals by a co-precipitation method for the synergistic removal of NO and CB. According to the findings, MnO_x and CeO₂ formed Mn_yCe_{1-y}O_{2-δ} solid solution. When the Mn content was 40%, the redox performance of the catalysts was well balanced with the surface acidity, resulting in catalysts having excellent activity in the reduction of NO and catalytic oxidation of CB in the range of 200–300 °C. In addition, to further improve the performance of the synergistic catalytic removal of NO_x and CB at low temperatures, Yang et al. [40] prepared Nb-doped MnCe_{0.2}O_x composite oxide catalysts by the homogeneous precipitation method. The results showed that the introduction of Nb increased the average pore size, pore volume, and specific surface area of MnCe_{0.2}O_x; enhanced the stability of the catalyst structure; and significantly promoted the growth of Lewis acid amount at 220 °C. The removal rates of NO_x and CB reached 94.5% and 96%, respectively, with excellent sulfur resistance and water resistance.

Fe is widely available, reasonably priced, and stable. However, the single component FeO_x has low activity in the synergistic removal of NO_x and CB, and the catalytic activity can be substantially improved by doping the composite. Yin et al. [39] integrated FeVO₄

and Fe₂O₃ semiconductor materials into a bifunctional catalyst and balanced the redox properties and surface acidity of the catalyst by interfacial charge modulation to ensure excellent NO_x and CB synergistic removal performance. The optimized FeVO₄-Fe₂O₃ catalyst showed stable bifunctional removal of NO_x and CB (>95%) and high HCl selectivity (>85%) over the state-of-the-art V₂O₅-WO₃/TiO₂ catalyst. This provides an effective strategy for the design of advanced catalysts for multi-pollutant control.

The activity of the catalyst is influenced by the valence of the doped metal. Doping with different metal elements can regulate the amount of Brønsted and Lewis acids on the catalyst. Brønsted acid can provide protons that facilitate the breaking of C-Cl bonds and promote the production of HCl [56]. The strength of Lewis acid determines the redox performance of the catalyst and promotes the deep oxidation of intermediates, thus enhancing CO₂ selectivity [57]. Wei et al. [58] doped CeO₂ with low-valence Al³⁺ and high-valence Ta⁵⁺, respectively. It was found that the introduction of low-valence Al³⁺ increased the number of Lewis acid sites in the catalyst and facilitated the formation of lattice oxygen, thus achieving excellent performance in the synergistic removal of NO and CB through characterization and simulation. In contrast, the introduction of the higher valence state Ta⁵⁺ brought the opposite effect. Ta⁵⁺ provided more electrons to the ligand oxygen than Ce⁴⁺, which hindered the detachment of lattice oxygen from the catalyst surface and weakened the oxidation ability of the catalyst, and thus performed poorly in NH₃-SCR and CBCO.

Table 2. Non-loaded transition metal oxide catalysts and their synergistic removal efficiencies for NO and CB.

Catalyst	Reaction Conditions	Conversion	Reference
FeVO ₄ -Fe ₂ O ₃	NO _x = 500 ppm, CB = 50 ppm, NH ₃ = 515 or 485 ppm, O ₂ = 10 vol%, H ₂ O = 5 vol%, SO ₂ = 100 ppm, N ₂ as balance gas GHSV = 60,000 h ⁻¹	NO _x : ~100% (200 °C) CB: ~90% (275 °C)	[39]
MnNb _{0.4} Ce _{0.2} O ₂	NO _x = 600 ppm, CB = 50 ppm, NH ₃ = 600 ppm, O ₂ = 12 vol%, H ₂ O = 7 vol%, N ₂ as balance gas GHSV = 30,000 h ⁻¹	NO _x : ~100% (170 °C) CB: ~90% (250 °C)	[40]
MnO _{x(0.4)} -CeO ₂	NO = 500 ppm, CB = 50 ppm, NH ₃ = 500 ppm, O ₂ = 10 vol%, N ₂ as balance gas GHSV = 60,000 h ⁻¹	NO _x : ~100% (200 °C) CB: ~90% (270 °C)	[55]
Al _{0.1} -CeO ₂	NO = 500 ppm, CB = 500 ppm, NH ₃ = 500 ppm, O ₂ = 10 vol%, H ₂ O = 5 vol%, N ₂ as balance gas GHSV = 40,000 h ⁻¹	NO _x : ~100% (300 °C) CB: ~90% (300 °C)	[58]
MnFe _{0.7}	NO = 500 ppm, NH ₃ = 550 ppm, O ₂ = 2 vol%, or CB = 250 ppm, O ₂ = 2 vol%, N ₂ as balance gas GHSV = 100,000 h ⁻¹	NO _x : >90% (150 °C) CB: ~40% (275 °C)	[59]
SO ₄ ²⁻ _{0.10} Fe-MnO _x	NO = 500 ppm, CB = 100 ppm, NH ₃ = 500 ppm, O ₂ = 3 vol%, N ₂ as balance gas GHSV = 30,000 h ⁻¹	NO _x : >90% (140 °C) CB: ~90% (200 °C)	[60]

2.2. Loaded Transition Metal Oxide Catalysts

Loaded transition metal oxide catalysts usually contain one or more transition metal oxides as active components, which are uniformly distributed on carriers such as TiO₂, SiO₂, CeO₂, Al₂O₃, and molecular sieves. Generally, the carrier materials have high specific surface area and good thermal stability, which help to improve the activity and durability of the catalyst. The catalyst surface structure is determined by the nature of the active component and the carrier itself, and the high dispersion of the active component on the carrier surface contributes to the improvement of catalytic activity. In addition, the success of catalytic removal of multi-pollutants relies heavily on the adsorption capacity of the multiphase interface for the target pollutants [61], and weakly acidic metal oxides such as TiO₂ and Al₂O₃ as carriers can provide abundant surface acid sites, which play an important role in NH₃ and CB adsorption. At the same time, the synergistic effect between the active component and the carrier is also beneficial for enhancing the reactivity of the catalyst. The efficiencies of some loaded transition metal oxide catalysts for the synergistic removal of NO and CB are listed in Table 3.

The charge transfer between the active component oxide and the reduced carrier (e.g., TiO₂, CeO₂) is advantageous for increasing the catalyst activity. However, in practical studies, pure CeO₂ is seldom used as a catalyst carrier for the synergistic removal of pollutants such as NO_x and CB, because CeO₂ easily reacts with Cl, leading to a decrease in catalytic activity. Martín et al. [62] prepared two oxide catalysts, MnO_x and CeO_x, for the synergistic removal of NO and 1,2-dichlorobenzene (o-DCB) by co-precipitation and impregnation methods, respectively. It was found that compared with MnO_x/CeO₂ prepared by the impregnation method, the catalyst prepared by co-precipitation has better activity because of the MnO_x-CeO₂ solid solution, which improved redox, acidity, and resistance to chlorine. Furthermore, Bertinchamps et al. [63] demonstrated experimentally that most of the active metal oxides can be well dispersed on the TiO₂ surface, while the dispersion on the SiO₂ and Al₂O₃ surfaces is relatively poor. Therefore, the catalysts with TiO₂ as the carrier tend to show better catalytic activity.

VO_x-based catalysts with TiO₂ as a carrier are mainly used as commercial SCR catalysts with high deNO_x activity and anti-poisoning properties [64]. In recent years, it has been found that VO_x/TiO₂ catalysts also have good stability and catalytic activity in the catalytic oxidation of chlorinated aromatic pollutants such as CB. For the synergistic removal of pollutants such as NO_x and CB, the structure of surface VO_x plays an important role. It is generally believed that monomeric vanadium species are more effective for the oxidation of CB, o-DCB, etc., while polymerized vanadium species are more advantageous for the catalytic reduction of NO [65,66]. Zhai et al. [67] further investigated the reaction properties of VO_x/TiO₂ for the catalytic removal of NO_x and CB using a combination of DFT calculations and experimental investigations. It was found that the main adsorption site of CB on monomeric vanadium species was the V-OH bond, and the main adsorption site on the polymerized vanadium species was the V-O bond. The monomeric vanadium species facilitated the conversion of Lewis V-O to Brønsted V-OH, thus providing sufficient H protons for HCl formation, while the polymerized vanadium species could effectively retain the redox cycle of V⁴⁺/V⁵⁺, yielding superior activity in the CBCO and SCR reactions. In addition, the loading of VO_x on TiO₂ carriers has a very important effect on catalyst activity and selectivity, with the first growth followed by a decrease in pollutant conversion with increasing VO_x loading [68].

Additives such as WO_x and MoO_x are commonly added to VO_x/TiO₂ catalysts to provide additional surface acid centers and to prevent the transformation or sintering of the TiO₂ carrier from the anatase to rutile phase. They can significantly improve the catalytic performance and stability of catalysts, influence the surface acidity and VO_x dispersion of catalysts, and broaden the active temperature range [69,70]. As MoO₃ has better reduction and higher Brønsted acid center strength than WO₃, Mo-V/Ti exhibited higher SCR and CB oxidation activity than Mo-W-V/Ti and W-V/Ti catalysts, achieving 100% NO conversion and more than 95% CB conversion at 300–400 °C [49]. Yu et al. [46]

used Al_2O_3 and SiO_2 to further dope the Mo-V/Ti catalysts and investigated the effect of doping on the simultaneous removal of NO_x and CB. Compared with SiO_2 , the doping of Al_2O_3 broadened the active temperature window and provided more surface acidity, reaching 100% NO conversion at 250–400 °C and nearly 10% higher CB conversion of Al-VMo/Ti than VMo/Ti in the range of 300–350 °C. In addition, Al-VMo/Ti had a stronger tolerance to SO_2 while reducing the amount of polychlorinated by-products.

In addition to VO_x -based catalysts, other metal oxide catalysts (e.g., MnO_x , FeO_x) with TiO_2 as the carrier also have more outstanding catalytic activity in the removal of chlorinated aromatic pollutants such as CB. Li et al. [71] prepared a series of $\text{MnO}_x/\text{TiO}_2$ and $\text{MnO}_x\text{-SnO}_x/\text{TiO}_2$ catalysts by the co-precipitation method and investigated their CBCO performance, which showed that they had excellent performance in the low-temperature range. The introduction of Sn inhibited the formation of chlorine-containing oxides of Mn elements and suppressed the volatilization loss of the core active components, thus significantly improving the stability of the $\text{MnO}_x/\text{TiO}_2$ catalysts. The catalytic efficiency of $\text{MnO}_x\text{-SnO}_x/\text{TiO}_2$ for CB remained above 97% after 100 h of continuous reaction at 225 °C. Khaleel et al. [72] prepared $\text{Fe}_2\text{O}_3/\text{TiO}_2$ by the impregnation method, which can achieve complete conversion of CB at 400 °C. The doping of Ca can lead to the reduction in pore size, increase in specific surface area, and grain size reduction of $\text{Fe}_2\text{O}_3/\text{TiO}_2$ catalysts, which is beneficial for the catalyst activity [73]. However, these metal oxide catalysts are yet to be investigated for the synergistic removal of NO_x and CB.

The main disadvantages of TiO_2 compared with other carriers (e.g., Al_2O_3 and SiO_2) are its high cost, small specific surface area, and poor thermal stability. In contrast, composite carriers can combine the advantages of different carriers. For example, $\text{TiO}_2\text{-Al}_2\text{O}_3$ and $\text{TiO}_2\text{-SiO}_2$ composite carriers can combine the advantages of Al_2O_3 and SiO_2 in terms of large specific surface area, inhibit the clustering of surface active components, slow down the deactivation of catalysts, and significantly enhance the redox ability of catalysts [74,75]. In addition, multiphase carrier-loaded multi-metal oxide catalysts have been used for the synergistic removal of NO_x and CB. Jin et al. [48] designed a series of W-Zr- $\text{O}_x/\text{Ti-Ce-Mn-O}_x$ (WZ/TCM) catalysts for the synergistic removal of NO_x and CB. Among them, the active component W-Zr- O_x mainly provided solid super acids; CeO_2 and MnO_2 enhanced the redox performance; and TiO_2 had a large specific surface area. These factors ensured the excellent performance of the WZ/TCM catalyst. At 350–500 °C, the NO conversion of WZ/TCM reached 100% and the oxidation activity of CB was higher than 85%, with high N_2 selectivity and excellent sulfur and water resistance. Similarly, Jin et al. [76] prepared $\text{WCeMnO}_x/\text{TiO}_2\text{-ZrO}_2$ catalysts to study their catalytic performance for the synergistic catalytic removal of NO, Hg^0 and synergistic catalytic removal of NO and CB. The catalyst performance was improved by solid phase structure control. MnWO_4 improved the solid acidity of the catalyst and enhanced the catalytic activity at high temperatures; the formation of $\text{Ce}_{0.75}\text{Zr}_{0.25}\text{O}_2$, Ce_2WO_6 , $\text{Ce}_2\text{Zr}_2\text{O}_7$, and $\text{Ce}_2\text{Ti}_2\text{O}_7$ improved the low-temperature catalytic activity.

In addition to the commonly used TiO_2 carriers, molecular sieve carriers usually have a large specific surface area, good stability, sufficient surface acidity, special morphology, and unique advantages in terms of adsorption of reactants and reaction diversity [77]. In the removal of chlorinated aromatic pollutants such as CB, the appropriate acidity facilitates the conversion of Cl species to inorganic chlorine species and to some extent avoids the production of polychlorinated organics [78]. Gallastegi-Villa et al. [79] prepared metal-loaded ZSM-5 catalysts with Cu, Fe, Mn, and V and investigated their ability to synergistically catalyze the removal of NO and o-DCB. The highest catalytic activity of Cu/ZSM-5 was observed based on the TOF value (number of reactant molecules converted per unit time per unit catalytic active site) of o-DCB oxidation at 150 °C and the stability test results of NO conversion at 300 °C. However, it is still difficult to apply practically due to the presence of more polychlorinated by-products.

Table 3. Loaded transition metal oxide catalysts and their synergistic removal efficiency of NO and CB.

Catalyst	Reaction Conditions	Conversion	Reference
WZrO _x /TiCeMnO _x	NO = 500 ppm, CB = 100 ppm, NH ₃ = 500 ppm, O ₂ = 10 vol%, N ₂ as balance gas GHSV = 30,000 h ⁻¹	NO _x : ~100% (250 °C) CB: ~100% (400 °C)	[76]
V/Ti	NO = 500 ppm, CB = 50 ppm, NH ₃ = 500 ppm, O ₂ = 3.5 vol%, N ₂ as balance gas GHSV = 60,000 h ⁻¹	NO _x : ~100% (250 °C) CB: ~20% (300 °C)	[80]
V-Ce/Ti	NO = 500 ppm, CB = 50 ppm, NH ₃ = 500 ppm, O ₂ = 3.5 vol%, N ₂ as balance gas GHSV = 60,000 h ⁻¹	NO _x : ~100% (300 °C) CB: ~20% (300 °C)	[80]
V-Mn/Ti	NO = 500 ppm, CB = 50 ppm, NH ₃ = 500 ppm, O ₂ = 3.5 vol%, N ₂ as balance gas GHSV = 60,000 h ⁻¹	NO _x : ~100% (300 °C) CB: ~5% (300 °C)	[80]
V-W/Ti	NO = 600 ppm, CB = 100 ppm, NH ₃ = 600 ppm, O ₂ = 5 vol%, N ₂ as balance gas GHSV = 40,000 h ⁻¹	NO _x : ~100% (275 °C) CB: ~100% (350 °C)	[81]
V-Mo/Ti	NO = 500 ppm, CB = 100 ppm, NH ₃ = 500 ppm, O ₂ = 10 vol%, N ₂ as balance gas GHSV = 30,000 h ⁻¹	NO _x : ~100% (250 °C) CB: ~100% (350 °C)	[81]
V-Mo/Ti	NO = 500 ppm, CB = 100 ppm, NH ₃ = 500 ppm, O ₂ = 10 vol%, N ₂ as balance gas GHSV = 30,000 h ⁻¹	NO _x : ~100% (200 °C) CB: ~100% (300 °C)	[82]
Pd-V/Ti	NO = 600 ppm, CB = 100 ppm, NH ₃ = 600 ppm, O ₂ = 10 vol%, N ₂ as balance gas GHSV = 30,000 h ⁻¹	NO _x : ~100% (250 °C) CB: ~100% (400 °C)	[83]

3. Mechanism of Synergistic Multi-Reactant Removal Interaction

In the synergistic NO_x/CB removal process, NH₃-SCR and CBCO inevitably interact with each other. However, the interaction between multiple reactants is not single facilitation or inhibition but depends on various factors such as reaction temperature, reactant concentration, and catalyst properties (surface acidity, redox properties, adsorption properties). The interaction between multiple reactants is complex, and an in-depth understanding of the interaction mechanism of multiple reactants and the influence of reaction conditions on the synergistic removal efficiency can help guide the design and preparation of NO and CB synergistic removal catalysts.

3.1. Effect of NH₃-SCR on CBCO

Aniline, nitrobenzene, and benzonitrile were detected in the by-products of NO_x/CB co-removal, indicating that a portion of NH₃-SCR reactants was involved in CB oxidation [82,84,85]. It is of great practical importance to clarify the effect of NH₃-SCR reactants on CB oxidation efficiency and to investigate in-depth the mechanism of NH₃-SCR effect on CB oxidation to improve the removal rate of CB in the combined removal process.

V₂O₅-MoO₃/TiO₂ is currently the best medium- and high-temperature SCR catalyst for NO removal efficiency [23]; so, many NO_x/CB synergistic removal studies have been carried out based on V₂O₅-MoO₃/TiO₂ catalysts. However, there is no uniform knowledge of whether there is synergy between NH₃-SCR and CBCO on V₂O₅-MoO₃/TiO₂ catalysts.

Some researchers have found the existence of synergistic NH_3 -SCR and CBCO removal over V-M-T catalysts. NH_3 -SCR promotes CB-catalyzed oxidation because NO and NO_2 act as oxidants to accelerate the oxidation cycle of VO_x species. NO is oxidized to NO_2 at WO_x or MoO_x active sites on V_2O_5 - WO_3 (MoO_3)/ TiO_2 . NO_2 oxidation performance is greater than oxygen, which accelerates the conversion of V^{4+} to V^{5+} . V^{5+} is the active substance for CB oxidation; so, the increase in V^{5+} accelerates CB conversion and improves CB conversion [86,87]. However, by comparing the CB removal process alone on V-M-T catalyst with the CB removal process in NO_x /CB synergy, some researchers have found that there is a certain inhibition of CB oxidation by NH_3 -SCR; this is caused by the competitive adsorption between NH_3 , NO, and CB [80,82]. To better demonstrate the inhibitory effect of NH_3 and NO on CB adsorption, Gan et al. performed a series of TPD experiments (Figure 3) [88]. When NO, NH_3 , and CB were present as a single gas, their adsorption capacities were $238 \mu\text{mol g}^{-1}$, $145 \mu\text{mol g}^{-1}$, and $43 \mu\text{mol g}^{-1}$, respectively, indicating that all three reactants could be adsorbed on the catalyst. The adsorption capacity of CB pretreated with $\text{NO}+\text{NH}_3$ decreased to $3.7 \mu\text{mol g}^{-1}$, indicating a significant inhibitory effect of $\text{NO}+\text{NH}_3$ on CB adsorption. CB was mainly adsorbed on the Brønsted acidic site of the catalyst and completed the dechlorination process. The introduction of NH_3 and NO occupied the acid site on the catalyst surface, leading to a shift in the conversion temperature of CB toward higher temperatures [82,86]. To further clarify the effect of NH_3 and NO on CB oxidation, Gao et al. investigated in detail the effect of NH_3 and NO on CB in an aerobic environment [89]. On V_2O_5 - MoO_3 / TiO_2 catalysts, NH_3 binded significantly stronger to the catalyst than CB, the adsorption of CB decreased significantly when NH_3 was present, and the CB conversion efficiency decreased. NO has little effect on the adsorption of CB. NO readily combines with O_2 to form NO_2 gas, which is more oxidizing than O_2 and contributes to CB oxidation. When NH_3 , NO, and CB were added to the reaction simultaneously, the CB oxidation was promoted by NH_3 -SCR on V_2O_5 - MoO_3 / TiO_2 catalyst. On the Mn-Ce catalyst, the same promotion of CB oxidation by SCR was observed in the low-temperature section (below 200°C) [90].

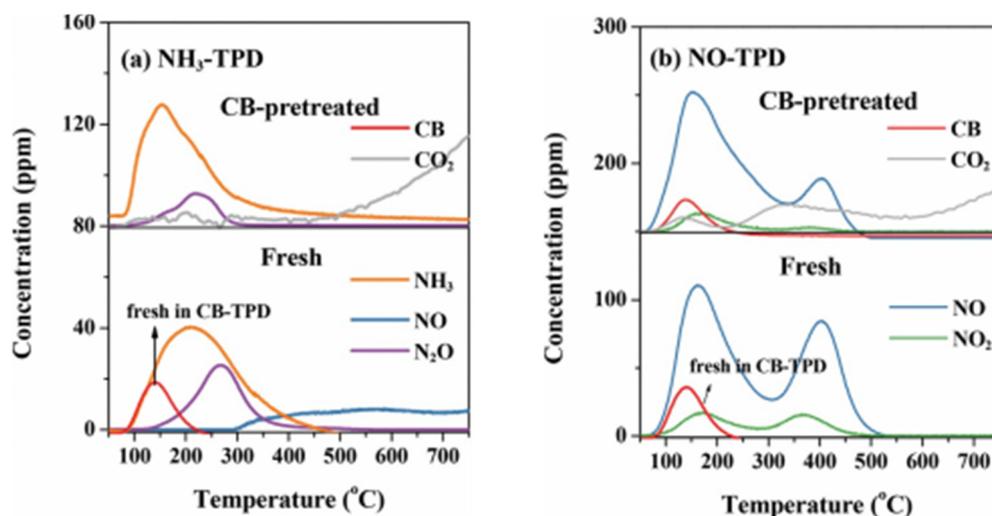


Figure 3. The NH_3 -TPD (a) and NO-TPD (b) profiles of the fresh and CB pre-treated catalysts. Reaction conditions: NO, 500 ppm (when used); CB, 50 ppm (when used); NH_3 , 500 ppm (when used); N_2 as the balance gas [88]. Copyright The Royal Society of Chemistry 2019.

There are obvious effects of gas components, intermediates, and reaction temperature on the oxidation of CB during the synergistic removal process. Li et al. prepared PdV/ TiO_2 and studied its synergistic NO and CB removal mechanism (Figure 4) [83]. In the range of 300 – 400°C , the catalytic removal efficiency of CB under different atmospheres was in the order of $\text{CBCO} + \text{NO}_2 > \text{CBCO} > \text{CBCO} + \text{NO} > \text{CBCO} + \text{NH}_3 > \text{CBCO} + \text{SCR}$. The O vacancies generated during the ring opening of CB facilitated the adsorption of the reacting

gas molecules and accelerated the CBCO + SCR reaction. The O vacancy structure on the catalyst surface is restored by NO_2 and O_2 reoxidation, and the presence of NO_2 molecules facilitates the CBCO reaction. More in-depth studies have shown that the promotion and inhibition of NO_2 on catalytic combustion of VOCs is related to its presence morphology. In the synergistic removal of NO and toluene, the effect of NO/ NO_2 / N_2O / O_2 content on the oxidation of toluene at 150 °C is shown in Figure 5. NO_2 cannot promote the oxidation of toluene when the NO_2 concentration is lower than 400 ppm; however, NO_2 has a promoting effect on the oxidation of toluene when the NO_2 content is higher than 400 ppm. This is because NO_2 forms nitrate species (NO_3^-) and nitrite species (NO_2^-) on the catalyst surface. Nitrate species (NO_3^-) are inert on the catalyst surface, do not have oxidation properties, and even cover the active site to affect the catalyst activity. As the concentration of NO_2 increases, nitrate production reaches equilibrium and NO_2 exists in adsorbed form. NO_2 in the adsorbed state has stronger oxidation properties than oxygen, thus promoting CB oxidation [85].

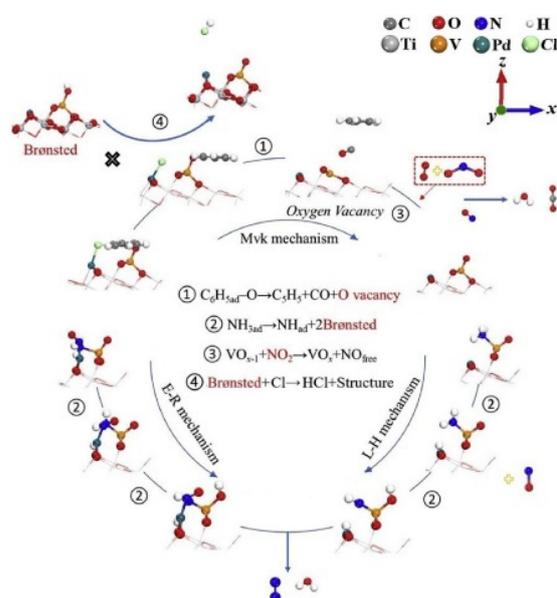


Figure 4. Synergistic removal mechanism of NO_x/CB on PdV/TiO_2 [83]. Copyright 2021, Elsevier B.V.

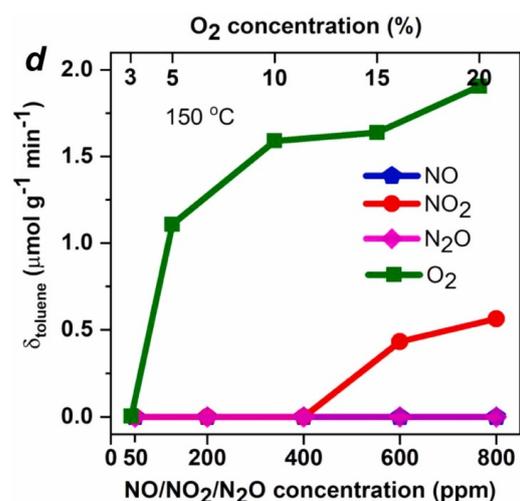


Figure 5. Effect of NO- NO_2 - N_2O - O_2 content on the oxidation of toluene at 150 °C [85]. Copyright 2021, Elsevier B.V.

3.2. Effect of CBCO on NH₃-SCR

Normally, the NO conversion efficiency in NH₃-SCR increases with the increase in temperature and remains stable in a certain temperature range when the NO conversion reaches its maximum; then, the NO conversion decreases as the C-O reaction occurs with the increase in temperature. The conversion efficiency of CB usually increases with the increase in temperature, reaches its maximum, and remains after reaching a certain temperature. In the NO_x/CB synergistic removal reaction, the initial temperature at which NO conversion occurs is usually lower than that of CB. Here, the effect of CB on NH₃-SCR is discussed in two parts: the low-temperature section (where CB is not oxidized) and the high-temperature section (where CB undergoes catalytic oxidation).

In the low-temperature section, CB is not decomposed and affects NH₃-SCR mainly by occupying the acidic sites with NO and NH₃. On most catalysts, the E-R mechanism coexists with the L-H mechanism during NO conversion; so, NO can participate in the reaction in the gaseous state without pre-adsorption on the catalyst. Therefore, the inhibition of NO adsorption by CB at low temperature has a negligible effect on NH₃-SCR. A series of temperature-programmed desorption (TPD) experiments showed a small decrease in NH₃ and NO adsorption after the adsorption of CB, and NH₃ adsorption was significantly higher than that of NO (Figure 4) [90]. In the NH₃-SCR reaction, NH₃ adsorbed at the L-acid site participates in the SCR reaction, while NH₃ adsorbed at the B-acid site is used as a supplement to the depletion of NH₃ at the L-acid site [88]. Thus, although CB leads to a decrease in NH₃ adsorption, the inhibitory effect on NO conversion is not significant.

In the high-temperature section, CB is catalytically converted to produce large amounts of CO₂, HCl, Cl₂, and some intermediate products (polychlorinated compounds). The C-Cl bond is broken during CB oxidation. The produced free Cl⁻ ions combine with the metal in the catalyst forming metal chlorides to activate neighboring bridging hydroxyl groups, providing additional B-acid sites, allowing for increased NH₃ adsorption, avoiding excessive oxidation of NH₃, and inhibiting the C-O oxidation reactions [88,89]. With the rise in NH₃ adsorption, the inactive NH₄Cl forms and occupies the catalyst's active site, regulating the redox performance of the catalyst and making the conversion efficiency of NO at higher temperatures. In the high-temperature section, CB can improve NO conversion efficiency and N₂ selectivity and broaden the SCR temperature window [48,88].

4. Catalyst Deactivation

Catalysts gradually decrease in catalytic activity and selectivity to deactivation during long-term use. Therefore, it is necessary to study the catalyst deactivation process and extend the catalyst life.

Chlorine poisoning and coking are the main causes of catalyst deactivation. The Cl⁻ generated by CB through nucleophilic or electrophilic substitution occupies the active site in the form of direct adsorption on the catalyst surface or occupying oxygen vacancies to form metal chlorides and metal chloride oxides. The generated NH₄Cl is deposited on the catalyst surface leading to catalyst deactivation. In the NO_x/CB synergistic removal process, the generation of by-products is another important factor in catalyst deactivation. The migration and transformation of carbon and chlorine during CB oxidation follow a dynamic equilibrium. Most of them are eventually converted into inorganic products CO_x and HCl or even Cl₂, some of which are present in the flue gas as organic matter and the rest accumulate on the catalyst surface as surface precipitates. CCl₄ and C₂Cl₄ are the main accumulators on the catalyst. They are the main by-products leading to catalyst deactivation [68].

The actual plant exhaust gas components are complex and contain various components such as SO₂, H₂O, HCl, and heavy metals. Among them, the reaction of SO₂ with NH₃ and H₂O result in the formation of (NH₄)₂SO₄ and NH₄HSO₄, which deposit on the catalyst surface and result in catalyst deactivation [91]. In addition, SO₂ can react with the catalyst's active component to form sulfides or sulfates that destroy or block the active sites on the catalyst surface. PdSO₄ and NH₄HSO₄ species are deposited on the catalyst surface and

occupy the catalyst active center, resulting in a blocked reaction on the catalyst surface and a decrease in catalytic efficiency. In general, sulfate poisoning such as $(\text{NH}_4)_2\text{SO}_4$ and NH_4HSO_4 is reversible. The generated sulfates (mainly NH_4HSO_4) are partially decomposed at $400\text{ }^\circ\text{C}$; then, the catalyst activity is recovered. In contrast, the deactivation caused by the formation of sulfate species from the catalyst active component with SO_2 is irreversible [92]. H_2O exerts an inhibitory effect on catalytic activity and this inhibition is generally reversible [93]. This inhibition is mainly caused by the competition of H_2O with the adsorption of the reactant gas on the surface of the active site. The degree of inhibition is influenced by the catalyst type, reaction temperature, and H_2O concentration, with higher H_2O concentrations showing more significant inhibition. In addition, H_2O is a necessary reactant for the formation of ammonium sulfate from NH_3 and SO_x ; so, the presence of H_2O aggravates the inhibitory effect of SO_2 on the catalyst activity. The inhibitory effect of HCl on the NH_3 -SCR reaction in low-temperature environments (below $250\text{ }^\circ\text{C}$) is evident [48]. This is attributed to the following two points: (1) the presence of HCl severely inhibits NH_3 adsorption; (2) NH_4Cl generated from HCl and NH_3 is deposited on the catalyst surface and covers the active site. Heavy metals in the flue gas also have a significant effect on the catalytic performance of the catalyst. As_2O_3 species can significantly reduce the formation energy of oxygen vacancies at the V-O-V sites on $\text{V}_2\text{O}_5\text{-WO}_3/\text{TiO}_2$ catalysts, which leads to the formation of more toxic polychlorinated by-products [81].

5. Conclusions

Both NH_3 -SCR and CBCO utilize the surface acidity and redox properties of the catalyst, which provides a theoretical basis for the synergistic removal of NO_x and CB. Balancing the surface acidity and redox properties of catalysts at a certain temperature is the key to the efficient removal of NO_x and CB. In this paper, the catalysts commonly used for the synergistic NO_x /CB removal are divided into two types: unloaded transition metal catalysts and loaded transition metal catalysts. Their catalytic properties are summarized and outlined, among which the unloaded transition metal catalysts are promising for their excellent catalytic performance. The inevitable interaction between NH_3 -SCR and CBCO in the synergistic NO_x /CB removal is discussed.

1. Effect of NH_3 -SCR on CBCO. NH_3 competes with CB for adsorption, and NH_3 has significantly stronger adsorption performance than CB, resulting in lower CBCO low-temperature activity. NO_2 is produced in the NH_3 -SCR process. NO_2 has a stronger oxidation performance than O_2 and can promote CB conversion. However, this promotion effect is related to the morphology of NO_2 , which can improve the CB conversion efficiency and reduce the CB ignition point when NO_2 is present in the adsorbed form in the reaction system.
2. Effect of CB on NH_3 -SCR. The effect of CB on the amount of NH_3 adsorbed was not significant; so, there was no significant inhibition of the NH_3 -SCR reaction by CB at low temperatures. The decomposition of CB at high-temperature and the generated Cl^- provided additional acid sites to increase the NH_3 adsorption and improve the NO conversion efficiency. The generated Cl^- reacts with NH_3 to form inert NH_4Cl , which regulates the redox performance of the catalyst, effectively inhibits the C-O reaction, and widens the SCR temperature window.

Catalyst deactivation is a difficult problem encountered in practical industrial applications. The cause of catalyst deactivation is the accumulation of carbon and chlorine species on the catalyst surface. The presence of SO_2 , H_2O , HCl , and heavy metals in complex flue gases leads to catalyst deactivation to different degrees. Rational design of catalysts; balancing the acidic and redox properties on the catalyst surface in a certain temperature interval; and improving N_2 , CO_2 selectivity, catalyst thermal stability, and resistance to poisoning are still bottlenecks that need to be broken in the future.

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