Preparation and Performance of Cerium-Based Catalysts for Selective Catalytic Reduction of Nitrogen Oxides: A Critical Review

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Abstract: Selective catalytic reduction of nitrogen oxides with NH3 (NH3-SCR) is still the most commonly used control technology for nitrogen oxides emission. Specifically, the application of rare earth materials has become more and more extensive. CeO2 was widely developed in NH3-SCR reaction due to its good redox performance, proper surface acidity and abundant resource reserves. Therefore, a large number of papers in the literature have described the research of cerium-based catalysts. This review critically summarized the development of the different components of cerium-based catalysts, and characterized the preparation methods, the catalytic performance and reaction mechanisms of the cerium-based catalysts for NH3-SCR. The purpose of this review is to highlight: (1) the modification effect of the various metal elements for cerium-based catalysts; (2) various synthesis methods of the cerium-based catalysts; and (3) the physicochemical properties of the various catalysts and clarify their relations to catalytic performances, particularly in the presence of SO2 and H2O. Finally, we hope that this work can give timely technical guidance and valuable insights for the applications of NH3-SCR in the field of NOx control.

Keywords: selective catalytic reduction; cerium-based catalysts; denitrification; physicochemical properties; catalytic performance

1. Introduction

NOx emissions from automobile exhausts and stationary sources pose a serious threat to environment. In 2017, the national NOx emission reached 17,852,200 tons, of which the NOx emissions from industrial sources was 6,459,000 tons, accounting for 36.2% of the total NOx emissions, and the NOx emissions from thermal power plants ranked first among the total NOx emissions from the key industrial enterprises under investigation [1]. Meanwhile, many countries have issued a number of laws and measures to strictly control NOx emissions, such as the New Sources Performance Standard of the United States, the Large Combustion Plant Directive: H 2001/80/EC of the European Union, the Air Pollution Prevention Law of Japan, the Atmospheric Environment Preservation Law of South Korea and the Thermal Power Plant of China (GB13223-2011).

The most promising approach to reduce NOx emissions is the selective catalytic reduction of NOx with NH3. The V2O5–WO3/TiO2 and V2O5–MoO3/TiO2 commercial catalysts were conventionally developed for NH3-SCR, because of their excellent catalytic performance and strong stability [2–5]. However, the poor catalytic temperature window (300–400 °C) and the toxicity of vanadium also bring difficulties for the disposal of the waste catalysts, which limit the future development of the vanadium-based catalysts [6,7]. Therefore, non-vanadium-based NH3-SCR catalysts currently attract significantly more attention in this field.

Apparently, China is the country with the most abundant rare earth mineral resources in the world, with not only with large reserves, but more importantly with complete mineral species and relatively low costs [8,9]. If rare earth oxide is applied to the research and
development of SCR catalysts, it can develop the high efficiency deNO\textsubscript{x} from industrial sources and automobile exhausts at low costs, which is the technical route for the preparation of SCR catalysts in accordance with China’s national conditions. Especially, CeO\textsubscript{2} plays a key role in the treatment of automobile exhausts, and also has certain significance for the abatement of particulate matter [10,11].

Up to now, CeO\textsubscript{2} as the main active component and promoter of NH\textsubscript{3}-SCR catalysts has been widely studied [12–14]. In general, CeO\textsubscript{2} is an acid-based substance, which has a large number of Lewis acid sites and a few Br\"{o}nsted acid sites. CeO\textsubscript{2} as an oxygen reservoir, which stores and releases oxygen via the redox shift between Ce\textsuperscript{4+} and Ce\textsuperscript{3+} under oxidizing and reducing conditions. Besides, CeO\textsubscript{2} exhibited an excellent SCR activity in the presence of SO\textsubscript{2} at 300–500 °C [15–18]. Furthermore, the most important properties of suitable surface acidity and good redox ability play a significant role in SCR performance [19,20]. Therefore, cerium-based catalysts were widely studied in NH\textsubscript{3}-SCR reaction [21,22]. In this paper, the research progress of cerium-based NH\textsubscript{3}-SCR catalysts made in recent years is summarized, including cerium-based bimetallic oxide catalysts, cerium-based multiplex oxide catalysts and cerium-based molecular sieve catalysts.

2. Cerium-Based Bimetallic Oxide Catalysts

CeO\textsubscript{2} enhances redox performance of the catalysts, which is vital for the catalytic reaction. Obviously, CeO\textsubscript{2} is responsible for the oxygen storage through the redox reaction, and Ce\textsuperscript{3+} increases the amount of unstable surface oxygen holes and oxygen free radicals [23–26]. However, the SCR performance of pure CeO\textsubscript{2} catalyst is poor, so many researchers have focused on synthesizing different composite catalysts for promoting NH\textsubscript{3}-SCR activity and extending the operating temperature windows. Therefore, the performance of cerium-based catalysts is continuously optimized by adding different metal oxides [27,28].

2.1. CeO\textsubscript{2}–TiO\textsubscript{2} Catalyst

Firstly, the CeO\textsubscript{2}–TiO\textsubscript{2} catalyst has been widely concerned due to good redox performance and its high specific surface area on the surface of catalyst [29,30]. As is known to all, TiO\textsubscript{2} is an optimal support of NH\textsubscript{3}-SCR catalysts with strong Lewis acidity and good SO\textsubscript{2} durability. Meanwhile, active components can be uniformly dispersed on its surface, consequently increasing the number of surface active sites [31,32]. Generally, the preparation methods of the CeO\textsubscript{2}–TiO\textsubscript{2} catalyst directly affect the strong interaction between CeO\textsubscript{2} and TiO\textsubscript{2} and the dispersion state of CeO\textsubscript{2} on the catalyst’s surface. The former mainly increases the specific surface. The latter directly affects the content of Ce\textsuperscript{3+} on the surface, thereby determining the redox performance of the catalysts. For example, Gao et al. [33,34] systematically compared the CeO\textsubscript{2}–TiO\textsubscript{2} catalysts obtained by impregnation method, sol–gel method and coprecipitation method. The results found that the catalyst prepared by the sol–gel method showed up to 93–98% NO\textsubscript{x} conversion at 300–400 °C. More specifically, the good deNO\textsubscript{x} performance might be attributed to the strong interaction between CeO\textsubscript{2} and TiO\textsubscript{2} shown in Figure 1. In addition, it can be also observed from Figure 2 that the primary particle size of CeTi (sol–gel) was less than 10 nm; meanwhile, these pictures revealed that CeO\textsubscript{2} was well dispersed on the surface of TiO\textsubscript{2}, thereby improving the catalytic activity of the catalysts. Similarly, Duan et al. [35] found that CeO\textsubscript{2} can be uniformly dispersed on the TiO\textsubscript{2}; moreover, the levels of CeO\textsubscript{2} in the catalyst did not change the crystal structure of the anatase. Obviously, the preparation methods of the sol–gel have great impact on the strong molecular interaction and surface characteristics of catalysts, which determine the NH\textsubscript{3}-SCR performance. Besides, Huang et al. [36] obtained a series of CeO\textsubscript{2}/TiO\textsubscript{2} catalysts with organic additives by the ball milling method. The results indicated that the addition of citric acid into the ball milling process could significantly change the proceedings of the precursor mixture decomposition, enhancing the dispersion and reducibility of the CeO\textsubscript{2} and the surface acidity as well as the surface microstructure.
milling method. The results indicated that the addition of citric acid into the ball milling process could significantly change the proceedings of the precursor mixture decomposition, enhancing the dispersion and reducibility of the CeO₂ and the surface acidity as well as the surface microstructure.

Figure 1. H₂-TPR profiles of the catalysts: (a) CeTi (sol–gel method), (b) CeTi (impregnation method), (c) CeTi (coprecipitation method) [33,34].

Furthermore, the presence of CeO₂ in the CeO₂–TiO₂ catalyst can effectively enhance the catalytic activity and thermostability of TiO₂. Especially, ceria atoms can inhibit the
crystalline grain growth and the collapse of the small channels generated by calcination [37]. In addition, some studies have found that the sulfate formed in the presence of SO2 is unstable on the surface of TiO2 and is easily decomposed; thus, TiO2 has high SO2 durability performance [38]. The function of the CeO2–TiO2 catalyst has been evaluated by Fei et al. [39]. Particularly, the Ce0.5Ti0.5 catalyst exhibited the best catalytic activity and extraordinary H2O/SO2 durability (Figures 3 and 4). Furthermore, the mechanism of NH3-SCR over CeaTi1−a catalysts was confirmed in Figure 5, where NH3, as the main active intermediate, reacted with NO to produce N2 and H2O in the E-R mechanism (Pathway 1). Besides, for the L-H mechanism, a large amount of Ce3+ species and high surface adsorbed oxygen reacted with adsorbed NH3 through the “fast SCR”, (Pathway 2). Subsequently, NO firstly adsorbed on the active sites and reacted with O− to form the intermediate, and lastly reacted with the adsorbed NH3 to form N2 and H2O (Pathway 3). Meanwhile, the effect of the loading sequence of CeO2 and TiO2 on the catalytic activity was investigated by Zhang et al. [40]. They also found that the TiO2/CeO2 catalyst not only showed good low-temperature activity at 150–250 °C, but also showed great SO2 resistance performance with the existence of 200 ppm SO2 at 300 °C. Actually, a large amount of CeO2 will actively react with SO2 in priority, avoiding the interaction between the SO2 and Ce–O–Ti active species; thereby the active species can completely exhibit great deNOx performance, as shown in Figure 6. Additionally, some investigators have done some work on the influence of different precursors of CeO2 and TiO2 on the catalysts’ performance. For instance, Yao et al. [41] synthesized CeO2/TiO2 catalyst with anatase, brookite and rutile TiO2 as support. The catalyst with rutile TiO2 exhibited great NH3-SCR activity owing to the large amount of acid sites, surface Ce3+ content, and surface adsorbed oxygen species. However, the H2O/SO2 durability performances of CeO2/TiO2 catalyst with rutile TiO2 need to be further studied and improved. The abovementioned reports suggested that CeO2 and TiO2 exhibited more acid sites and higher dispersion than the pure CeO2, which significantly enhanced the catalytic activity of the catalysts. SO2 and H2O showed a promotion on NOx reduction over Ce/TiO2 catalyst at higher temperature, whereas they show a great inhibitory effect at low temperature [42].

![Figure 3](image-url)  
*Figure 3.* NH3-SCR activity of TiO2, CeO2 and CeαTi1−a catalysts. Conditions: [NH3] = [NO] = 1000 ppm, [O2] = 3 vol.%, N2 as balance gas, total flow rate = 500 mL·min−1 [39].
2.2. CeO₂–MnO₂ Catalyst

Up to now, the Mn-based catalyst has been extensively investigated in the literature. It was found to have superior low temperature activity due to its rich variable valence states (MnO, Mn₃O₄, Mn₅O₈, Mn₂O₃, and MnO₂) and huge surface area [43–45]. The presence of the Mn⁴⁺ species and its redox process are important for the excellent performance of the catalyst.

![Figure 4. Effect of (a) SO₂, (b) H₂O and (c) SO₂ + H₂O on NH₃-SCR activity over Ce₀.5Ti₀.5 catalyst at 300 °C. Conditions: [NH₃] = [NO] = 1000 ppm, [SO₂] = 100 ppm, [H₂O] = 10 vol.%, [O₂] = 3 vol.%, N₂ as balance gas [39].](image)

![Figure 5. Proposed NH₃-SCR reaction mechanism over CeₐTi₁−ₐ catalysts [39].](image)

![Figure 6. Proposed adsorption model of SO₂: (a) CeO₂, (b) Ce/Ti and (c) Ti/Ce [40].](image)
2.2. CeO$_2$–MnO$_2$ Catalyst

Up to now, the Mn-based catalyst has been extensively investigated in the literature. It was found to have superior low temperature activity due to its rich variable valence states (MnO, Mn$_3$O$_4$, Mn$_5$O$_8$, Mn$_2$O$_3$, and MnO$_2$) and huge surface area [43–45]. The presence of the Mn$^{4+}$ species and its redox process are important for the excellent NH$_3$-SCR activity at low temperatures and for N$_2$ selectivity [46–49]. At the same time, the thermal stability, chemisorbed oxygen and the concentration of O$_\alpha$ species on the surface will be improved by MnO$_x$ and CeO$_2$ [50]. However, the obstacle to the application of Mn-based catalysts is the poor performance of resisting H$_2$O and SO$_2$ [51]. The addition of CeO$_2$ can enhance resistance to H$_2$O and SO$_2$ to a certain extent [52,53]. For example, Qi et al. [54,55] introduced the CeO$_2$–MnO$_x$, which catalyst showed great H$_2$O/SO$_2$ durability. Moreover, Mn ions entered the lattice of CeO$_2$ and a large number of chemisorbed oxygen species were released to the surface, thereby enhancing the NH$_3$-SCR activity of the catalysts.

Besides, Ce and Mn can present different valence states under suitable preparation methods and reaction conditions. Furthermore, the strong interaction between CeO$_2$ and MnO$_2$ will make the catalysts show excellent low-temperature activity and improve the redox performance of the catalysts. For example, the different preparation methods of the MnO$_x$–CeO$_2$ catalyst have been analyzed by Shen et al. [56]. It was demonstrated that the hydrolysis process method suggested higher SCR activity in the temperature range of 80–260 °C; meanwhile, this catalyst showed higher Mn$^{4+}$/Mn$^{3+}$, Ce$^{4+}$/Ce$^{3+}$ ratio, higher specific area and higher O$_\alpha$/O$_p$ ratio. Apart from some mature preparation methods, many researchers have made innovative works about preparation methods; Yao et al. [57] reported that the MnO$_x$–CeO$_2$ catalyst prepared by the hydrothermal treatment method revealed the best NH$_3$-SCR performance and good resistance to SO$_2$ and H$_2$O (Figure 7a,b). The XRD patterns and Raman spectra characterization were shown in Figure 7c,d, where it was demonstrated that CeO$_2$ and MnO$_x$ had a strong interaction under the conditions of high temperature and high pressure. Furthermore, Mn$^{3+}$ entered into the lattice of CeO$_2$ to form Mn–O–Ce solid solution, which enhanced the SCR performance of the catalysts. Andreoli et al. [58] prepared CeO$_2$–MnO$_x$ catalysts by the solution combustion synthesis method, and this catalyst exhibited a higher NO$_x$ conversion of more than 90% at 120–350 °C. Besides, Liu et al. [59] synthesized the MnO$_x$–CeO$_2$ catalyst by the surfactant-template (ST) method and coprecipitation (CP) method, and the XRD demonstrated that smaller mixed oxide particles were obtained by the surfactant-template method. The smaller particles could contribute to improving the SCR performance, as shown in Figure 8a. Meanwhile, more reducible subsurface and bulk oxygen were clearly observed in the H$_2$-TPR, as shown in Figure 8b. However, one problem relating to the Mn based catalyst is that its application at low temperature is a big challenge. The main problem is that the N$_2$ selectivity will decrease significantly at high temperature. Meanwhile, the catalytic activity will still be inhibited by H$_2$O and SO$_2$, which cannot meet the requirements of industrial production [60].
Figure 7. (a) NO conversion of the synthesized MnO\textsubscript{x}–CeO\textsubscript{2} catalysts; (b) the H\textsubscript{2}O resistance of MnCe-HTM catalyst at 200 °C; (c) XRD patterns and of the synthesized catalysts and; (d) Raman spectra of the synthesized catalysts [57].

Figure 8. (a) XRD patterns of Mn\textsubscript{5}Ce\textsubscript{5} (CP) (A) and Mn\textsubscript{5}Ce\textsubscript{5} (ST) (B) catalysts and (b) H\textsubscript{2}-TPR profiles for Mn\textsubscript{5}Ce\textsubscript{5} (CP) (A) and Mn\textsubscript{5}Ce\textsubscript{5} (ST) (B) catalysts [39].
2.3. Other CeO$_2$–MO$_x$ Catalysts

In addition to the above composite metal oxides, other metal oxides as the main active components in NH$_3$-SCR catalysts have been extensively studied for low–medium temperatures. For example, the addition of WO$_3$ [61,62], Sn$_2$O$_5$ [63], MoO$_3$ [64], CuO$_x$ [65] and NiO [66] into Ce-based catalysts can improve the redox performance, surface acidity and the adsorption of NH$_3$ on the catalyst surface. Meanwhile, the addition of ZrO$_2$ [67], CoO [68] can enhance the specific surface area, thermal stability and the resistance to H$_2$O/SO$_2$ of cerium-based catalysts. The modification of cerium-based catalysts by doping WO$_3$ showed excellent de-NO$_x$ performance. For instance, Wang et al. [61] reported that the WO$_3$ was deposited on CeO$_2$ nanoparticles. This catalyst also exhibited the highest SCR activity below 300 °C, excellent H$_2$O/SO$_2$ resistance and good NH$_3$ adsorption at 125–450 °C. Generally, the presence of WO$_3$ provided more surface lattice oxygen O$_{2-}$ and acid sites at lower temperatures, which benefits the catalytic activity for NH$_3$-SCR. At the same time, the CeO$_2$–WO$_3$ catalyst has been reported on by Liu et al. [62].

The results suggested that the presence of W provided more acid sites, thereby generating additional chemisorbed oxygen, weakly adsorbed oxygen species and concentrations of Ce and Ce$^{3+}$ on the surface of the catalyst, shown in Figure 9. Liu et al. [63] investigated the performance of a CeO$_2$–SnO$_2$ catalyst for NH$_3$-SCR. The results revealed that the high catalytic performance of this catalyst was attributed to the synergetic effect between Ce and Sn species, which enhanced the redox ability, the Lewis acidity and the adsorption and activation of NH$_3$ species, thereby contributing to improving the NH$_3$-SCR performance.

![Figure 9. XPS spectra of (a) Ce3d and (b) W4f over CeO$_2$–WO$_3$ catalysts [62].](image)

Besides, the catalytic performance of the CeO$_2$–MoO$_3$ catalyst has been investigated by Peng et al. [64]. This catalyst showed good NH$_3$-SCR performance. Moreover, the Ce atoms and amorphous MoO$_3$ structure provided a large number of Lewis acid sites and Brønsted acid sites on the catalyst surface. Atribak et al. [67] reported the performance of the CeO$_2$–ZrO$_x$ catalyst at high temperature, and the results indicated the addition of Zr provided excellent thermal stability and more specific surface area of the catalysts. Apart from the traditional CeO$_2$–MO$_x$ catalysts, the single-atom catalysts have also showed great potential in the NH$_3$-SCR. Especially, adding a second late-transition metal into cerium-based catalyst as single atom could have great potential in the automobile exhaust field [69,70].

3. Cerium-Based Multiplex Oxide Catalysts

Cerium-based multiplex oxide catalysts are particularly outstanding owing to making up for the shortcomings of some single or bimetallic catalysts on NH$_3$-SCR activity. For CeO$_2$/TiO$_2$ catalyst, such as Mn, W and Mo are introduced to further improve the redox performance, the surface acidity and the H$_2$O/SO$_2$ durability of the catalysts. Therefore,
the effects of cerium-based multiplex oxide catalysts on de-NOx performance were mainly studied from the aspects of preparation methods, preparation conditions and additive doping modification.

3.1. Ce–Mn/TiO2 Catalyst

Apparently, MnOx has many changeable valence states. Its oxides can be converted to each other, which shows excellent catalytic activity at low temperature [71–73]. Meanwhile, CeO2 can reduce the loss of specific surface area and pore volume during calcination, which improves the oxygen storage capacity and redox performance of the catalysts. Besides, the interaction between MnOx and CeO2 can form Mn–O–Ce solid solution, thereby improving the adsorption and activation properties of NH3 [74]. For example, Liu et al. [75] developed the Mn–Ce/TiO2 catalyst by hydrothermal method. It was also found that the environmentally benign Mn–Ce/TiO2 catalyst exhibited excellent NH3-SCR activity and good resistance to H2O and SO2 with a wide temperature window. Meanwhile, this result showed that the dual redox cycles (Mn4+ + Ce3+ ↔ Mn3+ + Ce4+, Mn4+ + Ti3+ ↔ Mn3+ + Ti4+) might play a key role in the catalytic reaction, which facilitated the adsorption and activation of NH3, as shown in Figure 10. The structure and properties of 8% Mn–Ce/TiO2-PILC catalyst has been analyzed by Shen et al. [76]. The catalyst suggested rich mesoporous structure and large specific surface area. More specifically, it could be demonstrated that Ce modified Mn–Ce/TiO2-PILC catalyst enhanced the dispersion of Mn on the surface.

Compared with CeO2–MnOx catalyst, the resistance to H2O and SO2 of the Ce-Mn/TiO2 catalyst has been enhanced to some extent [77]. CeO2 can significantly inhibit the deposition of (NH4)2SO4 and NH4HSO4 on the catalyst surface, which is the fundamental reason for the improvement of SO2/H2O resistance [78]. For instance, that SO2 poisoning and regeneration of the Mn–Ce/TiO2 catalyst have been reported by Sheng et al. [79]. This catalyst showed good resistance to SO2; however, the deactivation of the Mn–Ce/TiO2 poisoned by SO2 still occurred. Then, Peng et al. [80] reported the influence of Ce addition on the potassium poisoning of the MnOx/TiO2 catalyst, and found that K can reduce the surface acidity and reduction performance of the catalyst. However, the presence of CeO2 can provide a certain number of Lewis acid sites, shown in Figure 11a; meanwhile, CeO2 enhanced the reducibility of Mn/Ti and maintained the redox performance of the SCR catalysts after potassium poisoning, shown in Figure 11b.
Figure 11. (a) NH3-TPD profiles of fresh and poisoned catalysts in the range of 75–400 °C and (b) H2-TPR profiles of fresh and poisoned catalysts in the range of 150–700 °C [80].

Eventually, from the reported work on the Ce–Mn/TiO2 catalyst, it is not difficult to find that the rich variable valence states of Mn show excellent NH3-SCR activity at low temperature. However, its SCR performance resistance to H2O/SO2 still needs to be further strengthened.

3.2. Ce–W/TiO2 Catalyst

Apparently, different aspects of W modified cerium-based catalysts have been widely studied, and highly dispersed WO3 is beneficial to improve the catalytic effect of the whole catalyst [81,82]. WO3, as a stabilizer and promoter, significantly increased the specific surface area, Ce3+/Ce4+ ratio and surface acid sites of the catalysts, consequently enhancing the adsorbed oxygen on the surface and the activated oxygen species [83,84]. Firstly, Chen et al. [85] developed Ce/TiO2 and W–Ce/TiO2 catalysts by the impregnation method. They also found that W–Ce/TiO2 catalyst showed better de-NOx performance. As shown in Figure 12, the presence of W provided more acid sites on the catalyst surface, and accelerated the reaction between NH4NO3 and NO to achieve a superior low-temperature activity. Then, Guo et al. [86] found that the CeO2–WO3/TiO2 catalyst showed good catalytic activity. Pretreated TiO2 made the surface active substances have higher dispersion. The addition of WO3 also enhanced the surface acidity and surface chemisorption oxygen. Meanwhile, the influence of WO3 intervention on the catalytic performance of MnCeW/m-TiO2 catalyst has been investigated by Zha et al. [87]. This catalyst showed excellent deNOx performance and N2 selectivity under the conditions of wide temperature window and high space velocity. Particularly, in situ DRIFTS, as shown in Figure 13, it found that the addition of WO3 enhanced more Brønsted acid sites on the surface at high temperature.

Additionally, some researchers have reported some innovative preparation methods. For example, Katarzyna et al. [88] prepared WO3/CeOx–TiO2 catalyst by the flame-spray synthesis method, and Figure 14 suggests the interpretation of the mechanism of particle formation during flame-spray synthesis method. This method further strengthened the interaction of WO3, CeO2 and TiO2. Meanwhile, the presence of WO3 increased Ce5+ and surface acidity on the catalyst surface to a great extent. The highly dispersed WO3 enhanced the Ce–O–W reaction and Ce–O–Ti reaction, and consequently improved the performance of the NH3-SCR catalysts. Besides, the addition of WO3 improved the thermal stability of the catalysts at 550–600 °C [89], and a large cerium oxide phase and more TiO2 crystal formation can be avoided in the catalytic reaction process [90].
Generally, the ratio of CeO₂ and WO₃ has great effect on the redox ability and surface characteristics of Ce-W/TiO₂ catalysts, which also determines the NH₃-SCR performance. For instance, the Ce₀.２W₀.２Ti catalyst with Ce/W molar ratio of 1:1 has been synthesized by Shan et al. [91]. It also showed that the best NH₃-SCR catalytic performance and 100% N₂ selectivity; above 90% of NO conversion was maintained from 275 °C to 450 °C. Besides, in our previous research, we have done some work on the Ce-W/TiO₂ catalyst for the NH₃-SCR reaction and analyzed the influence of the active components CeO₂ and WO₃ content on the de-NOx performance of the catalysts, and found that the 30Ce4W/TiO₂ catalyst showed up to 90% NOx conversion at the widest temperature range of 310 °C. More specifically, the results show that a higher proportion of Ce⁴⁺, more chemisorption of oxygen and high specific surface area were key for the excellent NH₃-SCR activity of this catalyst [92].
3.3. Ce–Mo/TiO2 Catalyst

Undoubtedly, it is not difficult to find that the addition of Mo can remarkably improve the performance of SCR catalysts. Especially, CeO2 and MoO3 can be highly dispersed on the surface of TiO2 carrier. Furthermore, Mo doping increases the Ce³⁺ content, creates more abundant Brønsted acid sites, and increases the oxygen vacancy and adsorbed oxygen substances on the catalyst surface [93,94]. Additionally, the presence of MoO3 could effectively enhance the SO2 and H2O resistance of the catalysts at low temperature [95]. For example, Li et al. [96] prepared an Mo-doped MoO3/CeO2–TiO2 (MoO3/CT) catalyst. The catalyst showed good low temperature activity and excellent SO2/H2O resistance performance (Figure 15a,b). More specifically, the addition of MoO3 increased the Brønsted acid sites on the catalyst surface, shown in Figure 16. Then, the influence of MoO3 modified CeO2–TiO2 catalyst on the NH3-SCR performance was systematically investigated by Liu et al. [97], who suggested that having more Brønsted acid sites was conducive to the adsorption of NH3. Furthermore, MoO3 can inhibit the formation of sulfates; thereby the catalyst simultaneously showed excellent SO2/H2O resistance. Besides, Ye et al. [98] prepared CeO2–MoO3/TiO2 catalysts by different kinds of methods, and found that the catalyst prepared by sol–gel method exhibited the widest reaction temperature window of 250–475 °C.

![Figure 15](image1.png)

**Figure 15.** (a) NO conversions on these MoO3/CeO2–TiO2 (Mo/CT) catalysts with different Mo and (b) H2O + SO2 resistance of the CeO2–TiO2 (CT) and Mo/CT catalysts at 250 °C [96].

![Figure 16](image2.png)

**Figure 16.** NH3 adsorption in situ DRIFTS of (a) CeO2–TiO2 (CT) and (b) MoO3/CeO2–TiO2 (Mo/CT) catalyst [96].

Additionally, some researchers added active components to CeO2–MoO3/TiO2 catalysts to enhance the deNOx performance of the catalyst. For instance, the NH3-SCR performance of a new type of CeO2–MoO3–WO3/TiO2 catalyst has been reported by Jiang...
et al. [99]. The result exhibited that the NO conversion was 93.8–98.9% at a GHSV of 90,000 h\(^{-1}\) and a temperature window of 275–450 °C. The presence of WO\(_3\) and MoO\(_3\) increased the adsorption capacity of NH\(_3\), the redox performance, the amount of Ce\(^{3+}\) and the chemisorption of oxygen on the surface. At the same time, the interaction between CeO\(_2\), MoO\(_3\), WO\(_3\) and TiO\(_2\) might play an increasingly vital role in the improvement of catalytic performance. Zhang et al. [100] developed the catalytic performance of the CeFMoTiO\(_x\) catalyst, which not only exhibited higher than 90% NO conversion at 240–420 °C, but also presented superior H\(_2\)O/SO\(_2\) durability. The results demonstrated that the presence of MoO\(_3\) improved the dispersity of CeO\(_2\) on the catalyst surface. The introduction of F increased the oxygen vacancy, consequently improved the redox performance of CeO\(_2\). Meanwhile, the Ti–F bond played a key role in the SCR reaction. Eventually, the poisoning mechanism of As on CeO\(_2\)–MoO\(_3\)/TiO\(_2\) catalyst has been analyzed by Li et al. [101], as shown in Figure 17. The results exhibited that As\(_2\)O\(_3\) would directly weaken the specific surface area, surface acidity and redox performance. However, after the addition of Mo, the stronger interaction between Mo and As can alleviate the effects of surface CeO\(_2\) poisoning to a certain extent, so as to recover the redox performance and Brønsted acid sites of the CeO\(_2\)–MoO\(_3\)/TiO\(_2\) catalyst.

![Figure 17. The performance of the arsenic resistance on MoO\(_3\) doped CeO\(_2\)/TiO\(_2\) catalyst for SCR of NO\(_x\) with ammonia [101].](image)

### 3.4. Other Cerium-Based Multiplex Oxide Catalysts

In addition to the above multiplex oxide catalysts, many research papers have reported that CeO\(_2\) was combined with other transition metal oxides to form NH\(_3\)-SCR catalysts, such as Sn\(_2\)O\(_5\) [102], VO\(_x\) [103], CuO [104], Nb\(_2\)O\(_5\) [105], ZrO\(_2\) [106] and CoO [107]. They can simultaneously enhance the surface acidity, redox performance and SO\(_2\)/H\(_2\)O resistance of the SCR catalysts. For instance, Zhang et al. [97] prepared CeSnTiO\(_x\) catalysts by the solvothermal method. The results suggest that the Sn doped catalysts showed better low-temperature activity, exhibiting an extraordinarily wide operation window ranging from 180 to 460 °C. Meanwhile, the H\(_2\)-TPR and XPS spectra results verified that the addition of Sn\(_2\)O\(_5\) improved the interaction between CeO\(_2\) and SnO\(_2\) and the redox ability of the catalysts. Then, a novel V\(_2\)O\(_5\)/CeTiO\(_x\) catalyst was introduced by Lian et al. [103]. The addition of VO\(_x\) enhanced catalytic activity, N\(_2\) selectivity and the resistance to SO\(_2\) and H\(_2\)O. Then, Li et al. [104] studied Cu modified Ce/TiO\(_2\) catalyst, and found that the catalyst with a Cu/Ce molar ratio of 0.005 showed the best low-temperature activity and excellent SO\(_2\) resistance performance. By means of XRD, BET, Raman, XPS and NH\(_3\)-TPD, it was demonstrated that the presence of CuO increased the amount of the surface adsorbed oxygen and Ce\(^{3+}\) species and created more Brønsted acid sites on the catalyst surface. Furthermore, the in situ DRIFT results demonstrated that CuO doping enhanced the adsorption capacity of NH\(_3\).
Besides, Jawaher et al. [105] prepared Nb$_5$Ce$_{40}$/Ti$_{10}$ catalyst by sol–gel method, and found that the catalyst showed up to 95% NO$_x$ conversion at 200 °C. The addition of Nb strengthened the surface acidity. Meanwhile, the strong interaction between Ce and Ti to form the Ce–O–Ti solid solution and the high dispersion of Nb$_2$O$_5$ can improve the NH$_3$-SCR activity. However, the presence of Nb$_2$O$_5$ will greatly decrease the specific surface area of the catalysts. Then, Zr modified Ce–W/TiO$_x$ catalyst was analyzed by Zhao et al. [106]. The presence of Zr enhanced more acidic sites, oxygen vacancies and adsorbed oxygen species on the surface, which showed the best NH$_3$-SCR catalytic activity and thermal stability. Liu et al. [107] found that the Co–Ce/TiO$_2$ catalyst exhibited good low-temperature activity, widened the temperature window and reacted quickly under the mechanism of L-H and E-R. Furthermore, the different particle sizes of Co$^{2+}$ and Ce$^{4+}$ promoted the Ce$^{3+}$ ratio and surface adsorption oxygen. Besides, Li et al. [108] prepared Ho-doped Mn–Ce/TiO$_2$ catalyst by impregnation method. The results indicated that the catalyst with Ho/Ti of 0.1 presented excellent catalytic activity with the NO conversion of more than 90% at 140–220 °C (Figure 18a,b). The characterization results showed that Ho increased the specific surface area and led to higher levels of chemisorbed oxygen, as shown in Figure 19; meanwhile, the presence of Ho inhibited the sulfation on the surface to some extent.

![Figure 18. (a) Comparison of catalytic performance of different catalysts and (b) effect of SO$_2$ + H$_2$O on NO conversion over Mn$_{0.4}$Ce$_{0.07}$/TiO$_2$ and Mn$_{0.4}$Ce$_{0.07}$Ho$_{0.1}$/TiO$_2$ catalysts [108].](image-url)
et al. [118]. The catalyst showed excellent NH$_3$-SCR activity and N$_2$ selectivity, mainly due to the construction strategy of Fe–ZSM-5@CeO$_2$ to increase the redox performance and active oxygen species of the catalyst, shown in Figure 21. Carja et al. [119] studied the Mn–Ce/ZSM-5 catalyst and the results exhibited good NH$_3$-SCR activity in the presence of H$_2$O and SO$_2$. More importantly, the synergistic interaction of ZSM-5 and Ce, Mn promoted microporous-mesoporous characteristics and specific surface properties of catalysts. Besides, Liu et al. [120] reported CuCe$_{0.75}$Zr$_{0.25}$O$_y$/ZSM-5 catalyst. The reaction temperature window was widened to 175–468 °C. According to XRD and TEM results, the

Figure 19. SEM images of two catalysts (a) Mn$_{0.4}$Ce$_{0.07}$Ho$_{0.1}$/TiO$_2$, (b) Mn$_{0.4}$Ce$_{0.07}$/TiO$_2$, (c) Mn$_{0.4}$Ce$_{0.07}$Ho$_{0.1}$/TiO$_2$ and (d) Mn$_{0.4}$Ce$_{0.07}$/TiO$_2$ [108].

4. Molecular Sieve Catalysts

Apart from the ceria-based composite oxide SCR catalysts, the excellent activity and high N$_2$ selectivity of molecular sieves are also considered as the most promising SCR catalysts [109,110]. Especially, molecular sieve catalyst has strong stability, toxicity resistance and wide reaction temperature range [111]. Among the molecular sieve catalysts, ZSM-5, Beta, USY and other carriers exhibit good adsorption capacity, moderate surface acidity and flexible reaction temperature window. Peculiarly, ZSM-5 exhibits stable crystal structure, good specific surface area, abundant acid sites and great thermal stability [112–114]. For example, Krishna K et al. [115] prepared the Ce/ZSM-5 catalyst by ion exchange method. The results suggested that CeO$_2$ is closely bound to ZSM-5, which provided more active sites to transform NO$_x$. Then, Liu et al. [116] prepared CeO$_2$-modified Cu/ZSM-5 catalyst by a wetness impregnation method, and found the presence of CeO$_2$ enhanced the NH$_3$-SCR activity of the catalyst at low temperature. However, this catalyst had a poor catalytic performance at high temperature. Additionally, Dou [117] analyzed that the addition of Ce can inhibit the crystallization of Cu and increase the dispersion of active component, which made the catalyst show better de-NO$_x$ performance at 148–427 °C. The Fe–ZSM-5@CeO$_2$ catalyst has been investigated by Chen et al. [118]. The catalyst showed excellent NH$_3$-SCR activity and N$_2$ selectivity, mainly due to the construction strategy of Fe–ZSM-5@CeO$_2$ to increase the redox performance and active oxygen species of the catalyst, shown in Figure 20. Subsequently, the surface Ce$^{4+}$ and active oxygen species over Fe–ZSM-5@CeO$_2$ promoted the adsorption and activation of NO, shown in Figure 21. Carja et al. [119] studied the Mn–Ce/ZSM-5 catalyst and the results exhibited good NH$_3$-SCR activity in the presence of H$_2$O and SO$_2$. More importantly, the synergistic interaction of ZSM-5 and Ce, Mn promoted microporous-mesoporous characteristics and specific surface properties of catalysts. Besides, Liu et al. [120] reported CuCe$_{0.75}$Zr$_{0.25}$O$_y$/ZSM-5 catalyst. The reaction temperature window was widened to 175–468 °C. According to XRD and TEM results, the
The proposed mechanism of NH₃-SCR over Fe–ZSM-5@CeO₂ [118].

In addition, both the β and USY zeolite catalysts have been mentioned slightly less often than the ZSM-5 zeolite catalyst. However, there are still some valuable studies to be found; for example, Liu et al. [121] reported the coating of CeO₂ shells on the surface of MoFe/Beta catalyst, as shown in Figure 22, and found that the presence of the CeO₂ shells enhanced the resistance to SO₂ and H₂O and high thermal stability. This was mainly due to the fact that both chemisorbed oxygen species and specific surface area were increased after the coating of the CeO₂ shells (Figures 23 and 24). Then, Huang et al. [122] reported Mn–Ce catalysts with β, ZSM-5 and USY molecular sieves as carriers, respectively, by the impregnation method and studied the de-NOₓ performance of the catalysts at low temperature. The results showed that the three zeolite supported Mn–Ce catalysts have good low temperature activity, and the Mn–Ce/USY catalyst showed up to 90% NOₓ conversion at 107 °C. The MnOₓ is mainly distributed on the catalyst surface in an amorphous structure. Meanwhile, the weak acid on the catalyst surface played a major role in the reaction.

Figure 20. The PDA (Personal Digital Assistant)-assisted route for Fe-ZSM-5@CeO₂ preparation [118].

Figure 21. The proposed mechanism of NH₃-SCR over Fe–ZSM-5@CeO₂ [118].
Finally, it is worth considering that the addition of active components and promoters can increase redox property for cerium-based SCR catalysts. Furthermore, the oxidation reaction of the catalysts was enhanced. However, the oxidation of SO\textsubscript{2} was simultaneously increased in the catalytic reaction process, thereby resulting in the formation of sulfate on the surface and inhibiting the NH\textsubscript{3}-SCR activity of the catalysts. Therefore, the question of the resistance to SO\textsubscript{2}, H\textsubscript{2}O needs to be further investigated. Finally, for all the above types of catalysts, the denitration performance of the catalysts under different preparation methods and conditions was described in Table 1.
of the resistance to SO$_2$, H$_2$O needs to be further investigated. Finally, for all the above types of catalysts, the denitration performance of the catalysts under different preparation methods and conditions was described in Table 1.

### Table 1. The denitration performance of different catalysts.

| Catalysts               | Method                        | Temperature Window/ $^\circ$C | NO$_x$ Conversion/% | Gas Hourly Space Velocity (GHSV)/h$^{-1}$ | Refs. |
|-------------------------|-------------------------------|-------------------------------|---------------------|------------------------------------------|-------|
| CeO$_2$/TiO$_2$         | Sol–gel                       | 300–400                       | 93–98%              | 50,000 h$^{-1}$                         | [31,32]|
| CeO$_2$/TiO$_2$         | Dry ball milling              | 180                           | 84.6%               | GHSV of 30,000 h$^{-1}$                 | [34]  |
| MnO$_x$–CeO$_2$         | One-step hydrolysis process   | 180                           | Over 90%            | GHSV of 30,000 h$^{-1}$                 | [53]  |
| CeO$_2$–WO$_3$          | Two-step hydrothermal impregnation | 300–450                      | 100%                | GHSV of 60,000 h$^{-1}$                 | [58]  |
| CeO$_2$–SnO$_2$         | Hydrothermal                  | 280–425                       | Over 90%            | GHSV of 128,000 h$^{-1}$               | [60]  |
| Mn–Ce/TiO$_2$           | Hydrothermal                  | 150–350                       | Over 90%            | GHSV of 64,000 h$^{-1}$                 | [70]  |
| Ce–W/TiO$_2$            | Sol–gel precipitation         | 210–460                       | Over 90%            | GHSV of 150,000 h$^{-1}$               | [81]  |
| Ce–Mo/TiO$_2$           | Sol–gel                       | 250–475                       | Over 90%            | GHSV of 90,000 h$^{-1}$                | [94]  |
| MnCeW/TiO$_2$           | Impregnation                  | 140–340                       | Over 95%            | GHSV of 40,000 h$^{-1}$                | [83]  |
| Ce–Cu/ZSM-5             | Wet impregnation              | 210–320                       | Over 90%            | GHSV of 100,000 h$^{-1}$               | [112] |
| MoFe/Beta@CeO$_2$       | Wet impregnation              | 225–600                       | Over 90%            | GHSV of 50,000 h$^{-1}$                | [117] |

### 5. Conclusions and Perspectives

In conclusion, cerium-based catalysts have been deeply studied due to their high deNO$_x$ performances and low costs. The catalytic performance of cerium-based catalysts mainly depends on surface acidity, specific surface area, redox performance and resistance to H$_2$O and SO$_2$. The current study indicated the better catalytic performance of cerium-based bimetallic oxides than pure CeO$_2$ in NH$_3$-SCR. Furthermore, the multiplex oxide catalysts present a wider operation temperature widow and great low-temperature activity than the bimetallic oxide catalysts. This is attributed to the synergistic interaction between active components and promoters, the enhancement of the acid sites and the redox properties. Moreover, not only the addition of the other metal oxides can modify the performance of cerium-based catalysts, but different synthesis methods can also enhance the dispersion of the active species and the interaction of the different active components, the cerium-based bimetallic oxide catalysts, the cerium-based multiplex oxide catalysts and cerium-based molecular sieve catalysts are still the research directions in NH$_3$-SCR field in the future. Some researchers have done fruitful work in the fields of the synthesis method, modification and catalytic mechanism of cerium-based catalysts. Nevertheless, some aspects need to be further investigated. First of all, at low temperature, the performance of catalysts is still inhibited by H$_2$O and SO$_2$. Due to that, the improvement of the SO$_2$/H$_2$O resistance of cerium-based catalysts is still the main research direction. Secondly, traditional synthesis methods of catalysts need to be further studied and new synthesis methods need to be explored in order to expose more active sites on the catalyst surface and enhance the interaction between the active components. Furthermore, in order to provide the excellent performance of cerium-based catalysts, it is necessary to further achieve the optimal ratio of the active components. Additionally, for the cost of the catalysts, some metal oxides have high costs, which cause substantial obstacles to their actual production. Therefore, to ensure the excellent catalytic performance of cerium-based catalysts, the active components with low costs should be selected.

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