

Review

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 NH_3 (NH_3 -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. CeO_2 was widely developed in NH_3 -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 NH_3 -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 SO_2 and H_2O . Finally, we hope that this work can give timely technical guidance and valuable insights for the applications of NH_3 -SCR in the field of NO_x control.

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



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1. Introduction

NO_x emissions from automobile exhausts and stationary sources pose a serious threat to environment. In 2017, the national NO_x emission reached 17,852,200 tons, of which the NO_x emissions from industrial sources was 6,459,000 tons, accounting for 36.2% of the total NO_x emissions, and the NO_x emissions from thermal power plants ranked first among the total NO_x emissions from the key industrial enterprises under investigation [1]. Meanwhile, many countries have issued a number of laws and measures to strictly control NO_x 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 NO_x emissions is the selective catalytic reduction of NO_x with NH_3 . The $\text{V}_2\text{O}_5\text{-WO}_3/\text{TiO}_2$ and $\text{V}_2\text{O}_5\text{-MoO}_3/\text{TiO}_2$ commercial catalysts were conventionally developed for NH_3 -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 NH_3 -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_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, CeO₂ 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, CeO₂ as the main active component and promoter of NH₃-SCR catalysts has been widely studied [12–14]. In general, CeO₂ is an acid-based substance, which has a large number of Lewis acid sites and a few Brønsted acid sites. CeO₂ is as an oxygen reservoir, which stores and releases oxygen via the redox shift between Ce⁴⁺ and Ce³⁺ under oxidizing and reducing conditions. Besides, CeO₂ exhibited an excellent SCR activity in the presence of SO₂ 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 NH₃-SCR reaction [21,22]. In this paper, the research progress of cerium-based NH₃-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

CeO₂ enhances redox performance of the catalysts, which is vital for the catalytic reaction. Obviously, CeO₂ is responsible for the oxygen storage through the redox reaction, and Ce³⁺ increases the amount of unstable surface oxygen holes and oxygen free radicals [23–26]. However, the SCR performance of pure CeO₂ catalyst is poor, so many researchers have focused on synthesizing different composite catalysts for promoting NH₃-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. CeO₂-TiO₂ Catalyst

Firstly, the CeO₂-TiO₂ 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, TiO₂ is an optimal support of NH₃-SCR catalysts with strong Lewis acidity and good SO₂ 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 CeO₂-TiO₂ catalyst directly affect the strong interaction between CeO₂ and TiO₂ and the dispersion state of CeO₂ on the catalyst's surface. The former mainly increases the specific surface. The latter directly affects the content of Ce³⁺ on the surface, thereby determining the redox performance of the catalysts. For example, Gao et al. [33,34] systematically compared the CeO₂-TiO₂ 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_x conversion at 300–400 °C. More specifically, the good deNO_x performance might be attributed to the strong interaction between CeO₂ and TiO₂, 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 CeO₂ was well dispersed on the surface of TiO₂, thereby improving the catalytic activity of the catalysts. Similarly, Duan et al. [35] found that CeO₂ can be uniformly dispersed on the TiO₂; moreover, the levels of CeO₂ 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 NH₃-SCR performance. Besides, Huang et al. [36] obtained a series of CeO₂/TiO₂ 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 CeO₂ and the surface acidity as well as the surface microstructure.

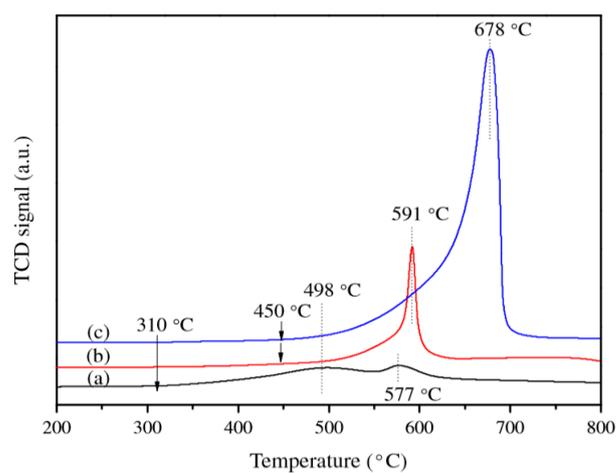


Figure 1. H_2 -TPR profiles of the catalysts: (a) CeTi (sol-gel method), (b) CeTi (impregnation method), (c) CeTi (coprecipitation method) [33,34].

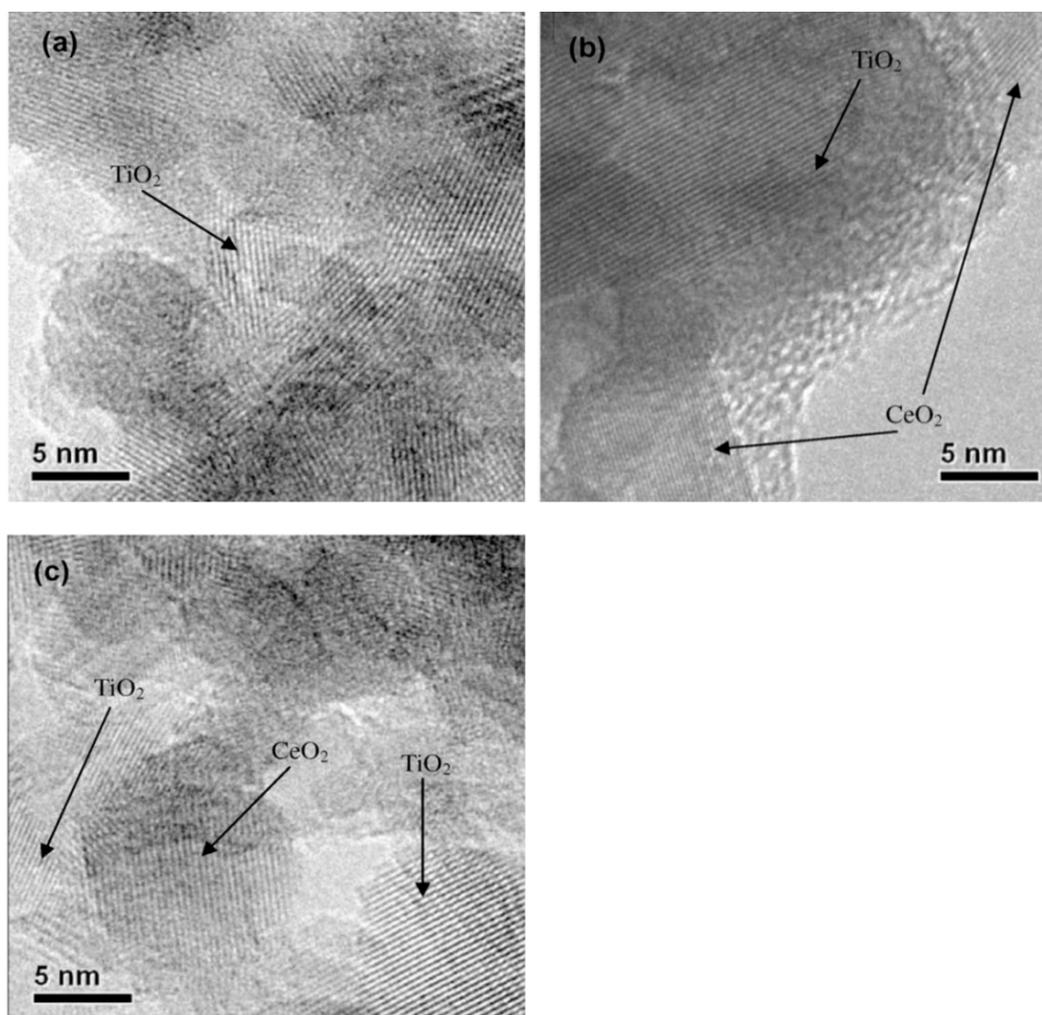


Figure 2. HR-TEM micrographs of the catalysts: (a) CeTi (sol-gel method), (b) CeTi (impregnation method) and (c) CeTi (coprecipitation method) [33,34].

Furthermore, the presence of CeO_2 in the CeO_2 - TiO_2 catalyst can effectively enhance the catalytic activity and thermostability of TiO_2 . 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 SO_2 is unstable on the surface of TiO_2 and is easily decomposed; thus, TiO_2 has high SO_2 durability performance [38]. The function of the $\text{CeO}_2\text{-TiO}_2$ catalyst has been evaluated by Fei et al. [39]. Particularly, the $\text{Ce}_{0.5}\text{Ti}_{0.5}$ catalyst exhibited the best catalytic activity and extraordinary $\text{H}_2\text{O}/\text{SO}_2$ durability (Figures 3 and 4). Furthermore, the mechanism of $\text{NH}_3\text{-SCR}$ over $\text{Ce}_a\text{Ti}_{1-a}$ catalysts was confirmed in Figure 5, where NH_3 , as the main active intermediate, reacted with NO to produce N_2 and H_2O in the E-R mechanism (Pathway 1). Besides, for the L-H mechanism, a large amount of Ce^{3+} species and high surface adsorbed oxygen reacted with adsorbed NH_3 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 NH_3 to form N_2 and H_2O (Pathway 3). Meanwhile, the effect of the loading sequence of CeO_2 and TiO_2 on the catalytic activity was investigated by Zhang et al. [40]. They also found that the $\text{TiO}_2/\text{CeO}_2$ catalyst not only showed good low-temperature activity at $150\text{--}250\text{ }^\circ\text{C}$, but also showed great SO_2 resistance performance with the existence of 200 ppm SO_2 at $300\text{ }^\circ\text{C}$. Actually, a large amount of CeO_2 will actively react with SO_2 in priority, avoiding the interaction between the SO_2 and Ce-O-Ti active species; thereby the active species can completely exhibit great deNO_x performance, as shown in Figure 6. Additionally, some investigators have done some work on the influence of different precursors of CeO_2 and TiO_2 on the catalysts' performance. For instance, Yao et al. [41] synthesized $\text{CeO}_2/\text{TiO}_2$ catalyst with anatase, brookite and rutile TiO_2 as support. The catalyst with rutile TiO_2 exhibited great $\text{NH}_3\text{-SCR}$ activity owing to the large amount of acid sites, surface Ce^{3+} content, and surface adsorbed oxygen species. However, the $\text{H}_2\text{O}/\text{SO}_2$ durability performances of $\text{CeO}_2/\text{TiO}_2$ catalyst with rutile TiO_2 need to be further studied and improved. The abovementioned reports suggested that CeO_2 and TiO_2 exhibited more acid sites and higher dispersion than the pure CeO_2 , which significantly enhanced the catalytic activity of the catalysts. SO_2 and H_2O showed a promotion on NO_x reduction over Ce/TiO_2 catalyst at higher temperature, whereas they show a great inhibitory effect at low temperature [42].

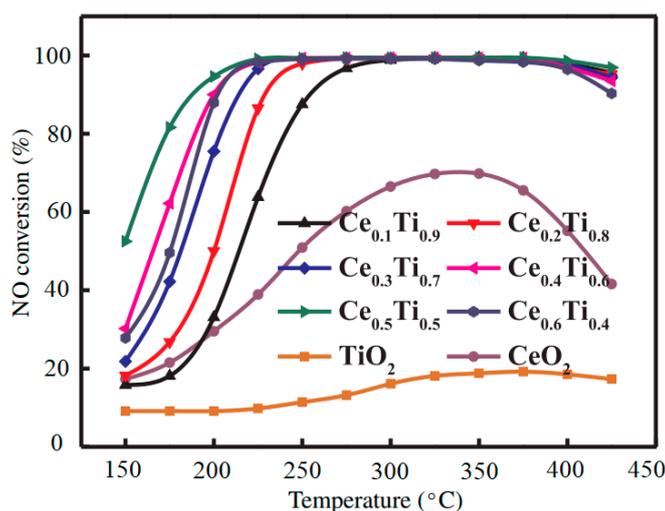


Figure 3. $\text{NH}_3\text{-SCR}$ activity of TiO_2 , CeO_2 and $\text{Ce}_a\text{Ti}_{1-a}$ catalysts. Conditions: $[\text{NH}_3] = [\text{NO}] = 1000\text{ ppm}$, $[\text{O}_2] = 3\text{ vol.}\%$, N_2 as balance gas, total flow rate = $500\text{ mL}\cdot\text{min}^{-1}$ [39].

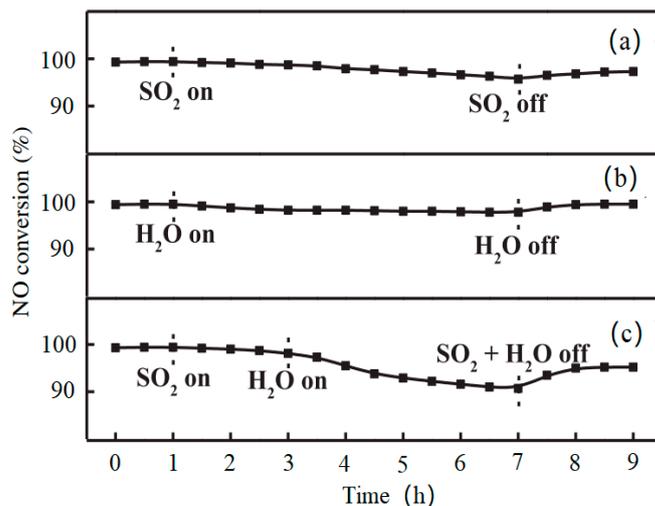


Figure 4. Effect of (a) SO₂, (b) H₂O and (c) SO₂ + H₂O on NH₃-SCR activity over Ce_{0.5}Ti_{0.5} catalyst at 300 °C. Conditions: [NH₃] = [NO] = 1000 ppm, [SO₂] = 100 ppm, [H₂O] = 10 vol.%, [O₂] = 3 vol.%, N₂ as balance gas [39].

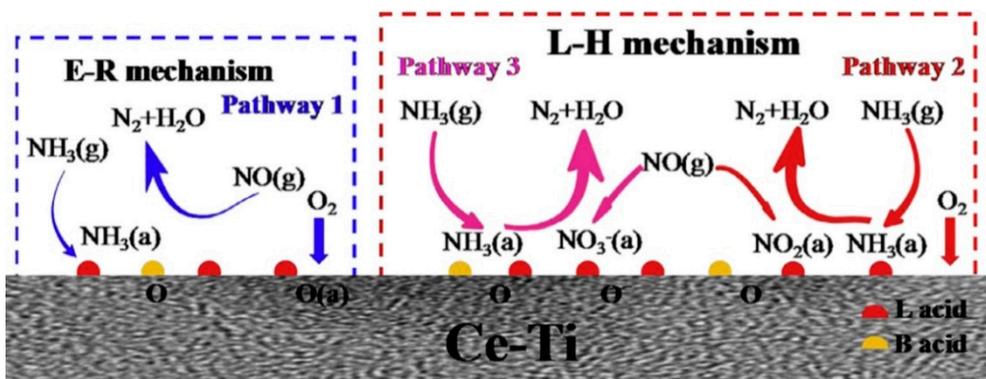


Figure 5. Proposed NH₃-SCR reaction mechanism over Ce_aTi_{1-a} catalysts [39].

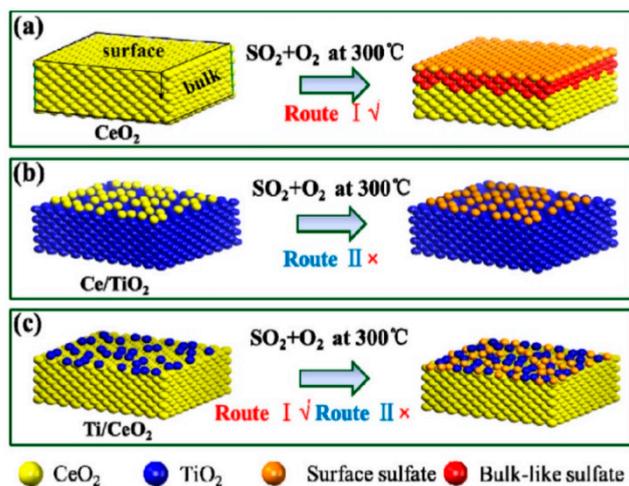


Figure 6. Proposed adsorption model of SO₂: (a) CeO₂, (b) Ce/Ti and (c) Ti/Ce [40].

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 NH₃-SCR activity at low temperatures and for N₂ selectivity [46–49]. At the same time, the thermal stability, chemisorbed oxygen and the concentration of O_α species on the surface will be improved by MnO_x and CeO₂ [50]. However, the obstacle to the application of Mn-based catalysts is the poor performance of resisting H₂O and SO₂ [51]. The addition of CeO₂ can enhance resistance to H₂O and SO₂ to a certain extent [52,53]. For example, Qi et al. [54,55] introduced the CeO₂–MnO_x, which catalyst showed great H₂O/SO₂ durability. Moreover, Mn ions entered the lattice of CeO₂ and a large number of chemisorbed oxygen species were released to the surface, thereby enhancing the NH₃-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 CeO₂ and MnO₂ 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–CeO₂ 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 Mn⁴⁺/Mn³⁺, Ce⁴⁺/Ce³⁺ ratio, higher specific area and higher O_a/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–CeO₂ catalyst prepared by the hydrothermal treatment method revealed the best NH₃-SCR performance and good resistance to SO₂ and H₂O (Figure 7a,b). The XRD patterns and Raman spectra characterization were shown in Figure 7c,d, where it was demonstrated that CeO₂ and MnO_x had a strong interaction under the conditions of high temperature and high pressure. Furthermore, Mnⁿ⁺ entered into the lattice of CeO₂ to form Mn–O–Ce solid solution, which enhanced the SCR performance of the catalysts. Andreoli et al. [58] prepared CeO₂–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–CeO₂ 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 H₂-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 N₂ selectivity will decrease significantly at high temperature. Meanwhile, the catalytic activity will still be inhibited by H₂O and SO₂, which cannot meet the requirements of industrial production [60].

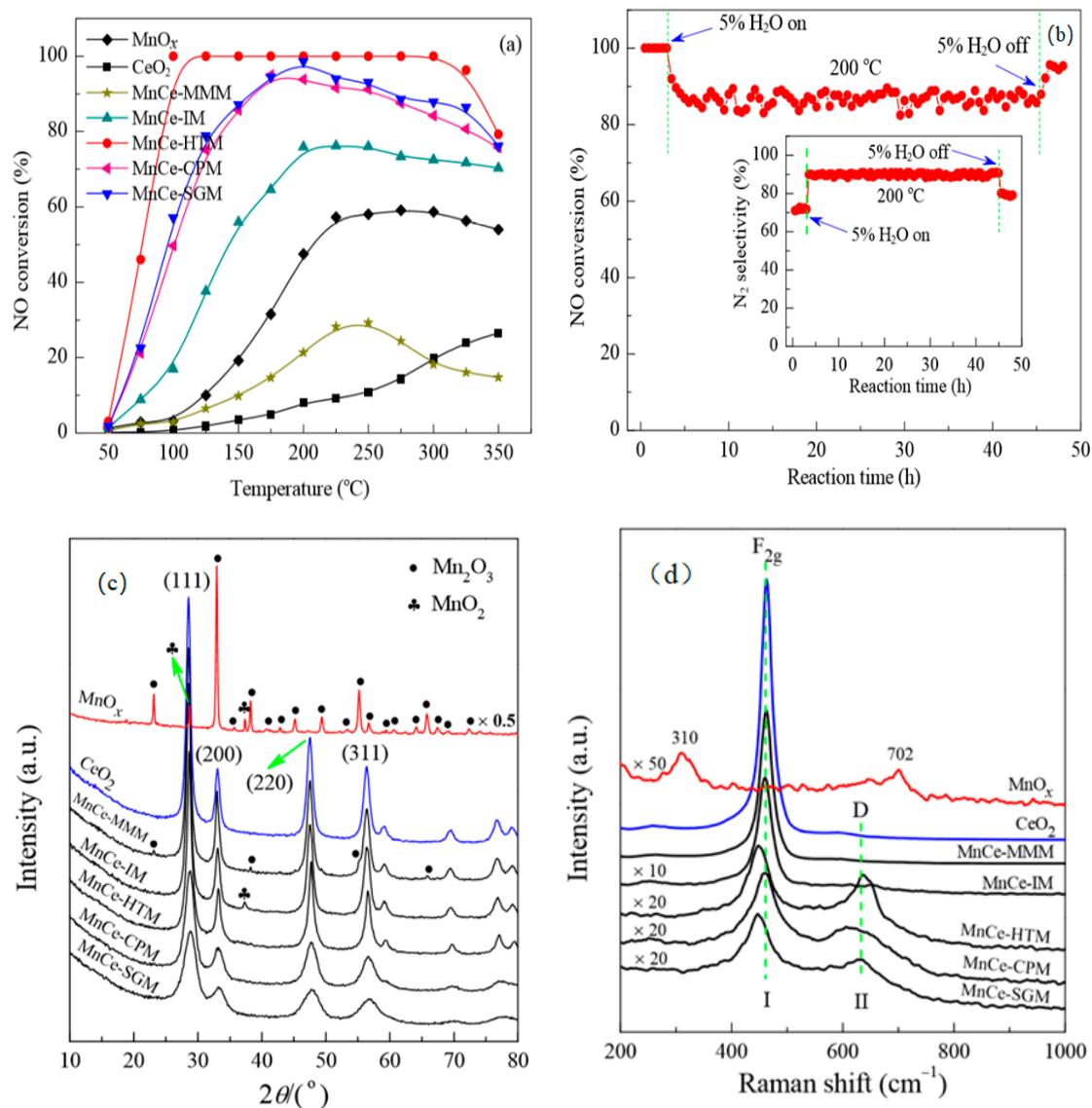


Figure 7. (a) NO conversion of the synthesized MnO_x-CeO₂ catalysts; (b) the H₂O resistance of MnCe-HTM catalyst at 200 °C; (c) XRD patterns and of the synthesized catalysts and; (d) Raman spectra of the synthesized catalysts [57].

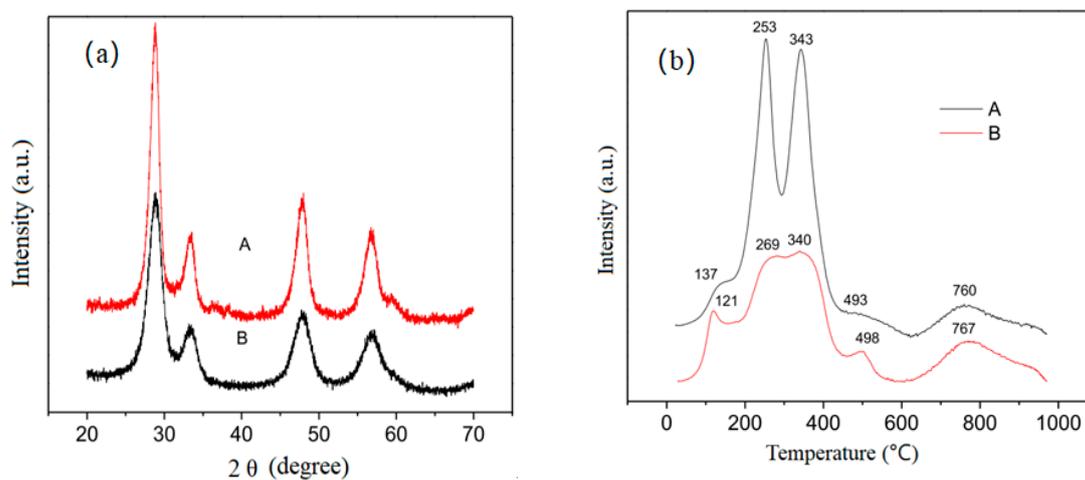


Figure 8. (a) XRD patterns of Mn₅Ce₅ (CP) (A) and Mn₅Ce₅ (ST) (B) catalysts and (b) H₂-TPR profiles for Mn₅Ce₅ (CP) (A) and Mn₅Ce₅ (ST) (B) catalysts [59].

2.3. Other CeO₂–MO_x Catalysts

In addition to the above composite metal oxides, other metal oxides as the main active components in NH₃-SCR catalysts have been extensively studied for low–medium temperatures. For example, the addition of WO₃ [61,62], Sn₂O₅ [63], MoO₃ [64], CuO_x [65] and NiO [66] into Ce-based catalysts can improve the redox performance, surface acidity and the adsorption of NH₃ on the catalyst surface. Meanwhile, the addition of ZrO₂ [67], CoO [68] can enhance the specific surface area, thermal stability and the resistance to H₂O/SO₂ of cerium-based catalysts. The modification of cerium-based catalysts by doping WO₃ showed excellent de-NO_x performance. For instance, Wang et al. [61] reported that the WO₃ was deposited on CeO₂ nanoparticles. This catalyst also exhibited the highest SCR activity below 300 °C, excellent H₂O/SO₂ resistance and good NH₃ adsorption at 125–450 °C. Generally, the presence of WO₃ provided more surface lattice oxygen O²⁻ and acid sites at lower temperatures, which benefits the catalytic activity for NH₃-SCR. At the same time, the CeO₂–WO₃ 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 Ce³⁺ on the surface of the catalyst, shown in Figure 9. Liu et al. [63] investigated the performance of a CeO₂–SnO₂ catalyst for NH₃-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 NH₃ species, thereby contributing to improving the NH₃-SCR performance.

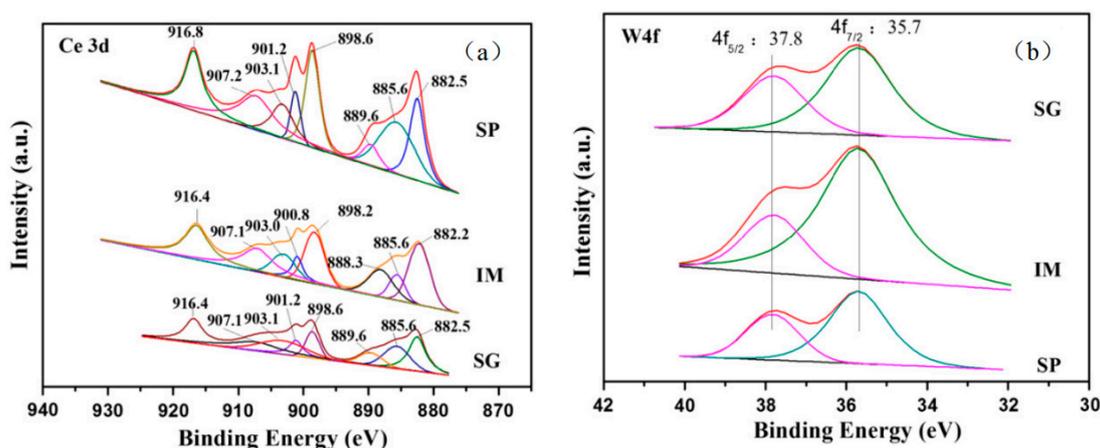


Figure 9. XPS spectra of (a) Ce3d and (b) W4f over CeO₂–WO₃ catalysts [62].

Besides, the catalytic performance of the CeO₂–MoO₃ catalyst has been investigated by Peng et al. [64]. This catalyst showed good NH₃-SCR performance. Moreover, the Ce atoms and amorphous MoO₃ 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 CeO₂–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 CeO₂–MO_x catalysts, the single-atom catalysts have also showed great potential in the NH₃-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 NH₃-SCR activity. For CeO₂/TiO₂ catalyst, such as Mn, W and Mo are introduced to further improve the redox performance, the surface acidity and the H₂O/SO₂ durability of the catalysts. Therefore,

the effects of cerium-based multiplex oxide catalysts on de-NO_x performance were mainly studied from the aspects of preparation methods, preparation conditions and additive doping modification.

3.1. Ce–Mn/TiO₂ Catalyst

Apparently, MnO_x 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 MnO_x 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.

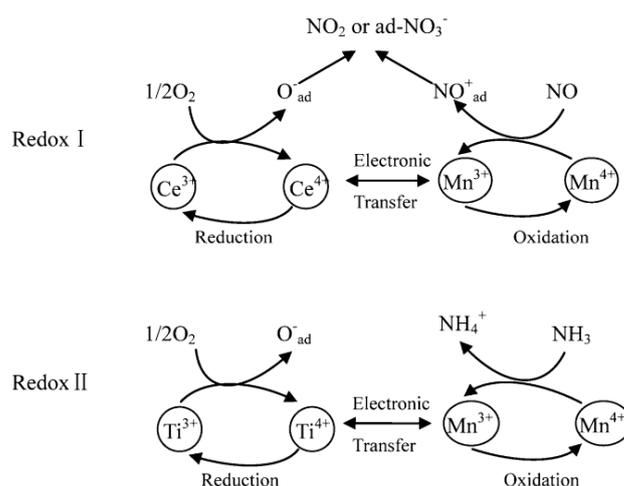


Figure 10. Dual redox cycles for the activation of NO and NH₃ [70].

Compared with CeO₂–MnO_x 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 MnO_x/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.

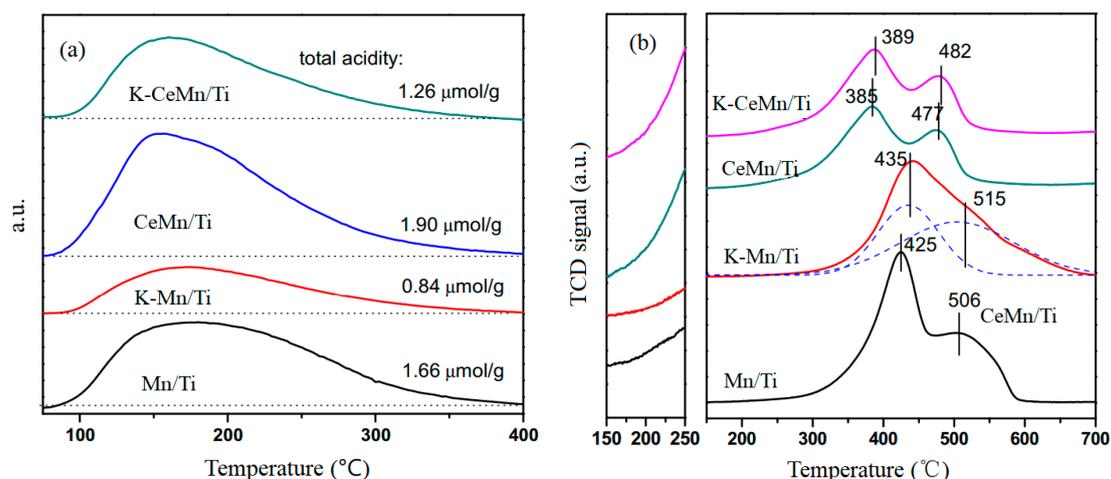


Figure 11. (a) NH₃-TPD profiles of fresh and poisoned catalysts in the range of 75–400 °C and (b) H₂-TPR profiles of fresh and poisoned catalysts in the range of 150–700 °C [80].

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-NO_x 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 deNO_x 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 Brönsted 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/CeO_x–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 Ce³⁺ 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].

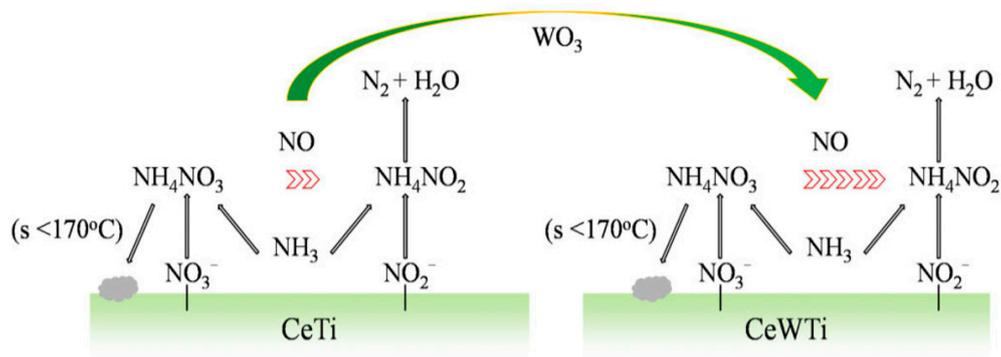


Figure 12. NH_3 -NO/ NO_2 SCR reaction routes on CeTi and CeWTi catalysts [85].

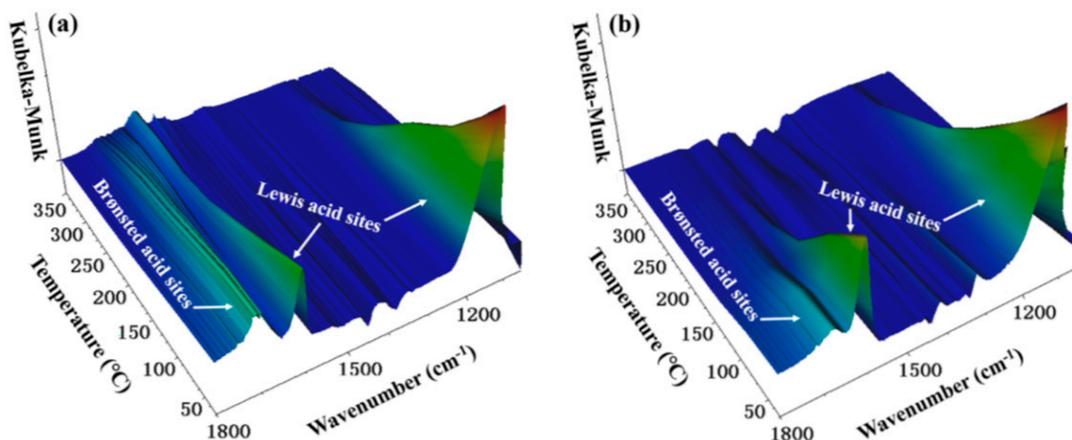


Figure 13. In situ DRIFTS of NH_3 desorption over (a) MnCeW/m- TiO_2 and (b) MnCe/m- TiO_2 catalysts as a function of temperature [87].

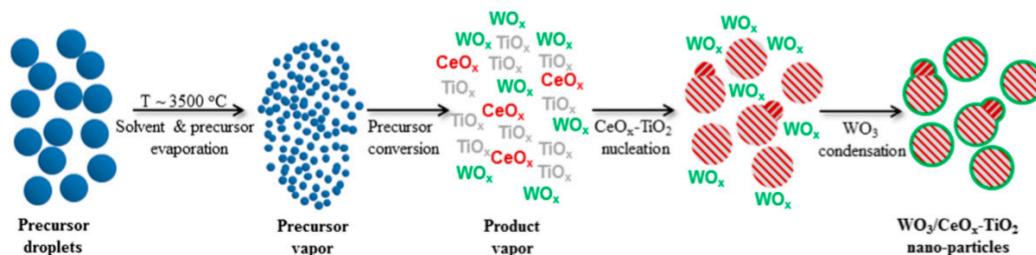


Figure 14. Schematic representation of a feasible mechanism of $\text{WO}_3/\text{CeO}_x\text{-TiO}_2$ nanoparticles formation during flame-spray synthesis [88].

Generally, the ratio of CeO_2 and WO_3 has great effect on the redox ability and surface characteristics of Ce-W/ TiO_2 catalysts, which also determines the NH_3 -SCR performance. For instance, the $\text{Ce}_{0.2}\text{W}_{0.2}\text{Ti}$ catalyst with Ce/W molar ratio of 1:1 has been synthesized by Shan et al. [91]. It also showed that the best NH_3 -SCR catalytic performance and 100% N_2 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/ TiO_2 catalyst for the NH_3 -SCR reaction and analyzed the influence of the active components CeO_2 and WO_3 content on the de- NO_x performance of the catalysts, and found that the 30Ce4W/ TiO_2 catalyst showed up to 90% NO_x conversion at the widest temperature range of 310 °C. More specifically, the results show that a higher proportion of Ce^{4+} , more chemisorption of oxygen and high specific surface area were key for the excellent NH_3 -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 Brönsted 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 (Mo₃/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 Brönsted 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 Brönsted 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.

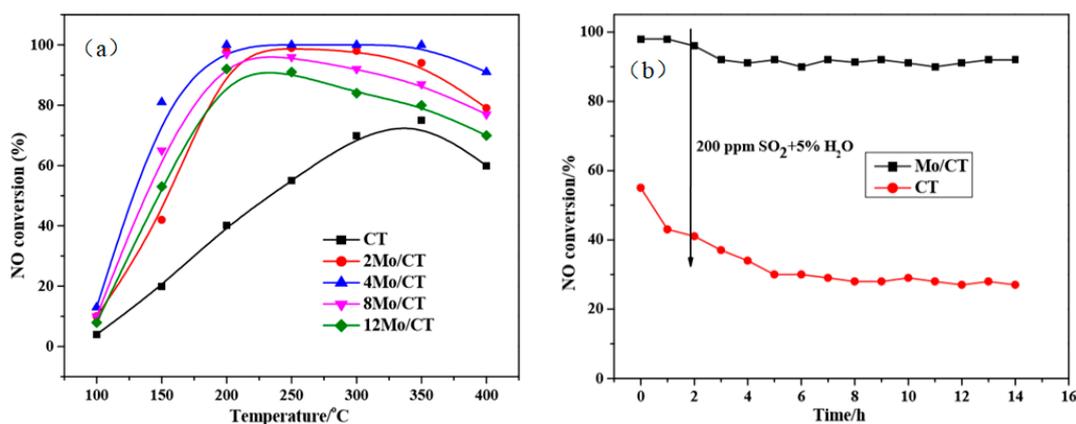


Figure 15. (a) NO conversions on these MoO₃/CeO₂-TiO₂ (Mo/CT) catalysts with different Mo and (b) H₂O + SO₂ resistance of the CeO₂-TiO₂ (CT) and Mo/CT catalysts at 250 °C [96].

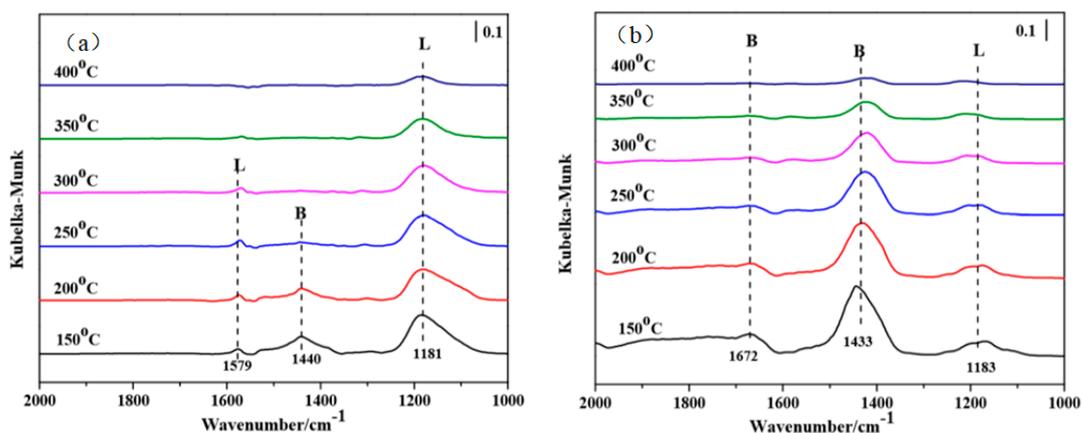


Figure 16. NH₃ adsorption in situ DRIFTS of (a) CeO₂-TiO₂ (CT) and (b) MoO₃/CeO₂-TiO₂ (Mo/CT) catalyst [96].

Additionally, some researchers added active components to CeO₂-MoO₃/TiO₂ catalysts to enhance the deNO_x 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 $90,000 \text{ h}^{-1}$ 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 CeFmTiO_x catalyst, which not only exhibited higher than 90% NO conversion at 240–420 °C, but also presented superior $\text{H}_2\text{O}/\text{SO}_2$ 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 $\text{CeO}_2\text{–MoO}_3/\text{TiO}_2$ catalyst has been analyzed by Li et al. [101], as shown in Figure 17. The results exhibited that As_2O_5 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 $\text{CeO}_2\text{–MoO}_3/\text{TiO}_2$ catalyst.

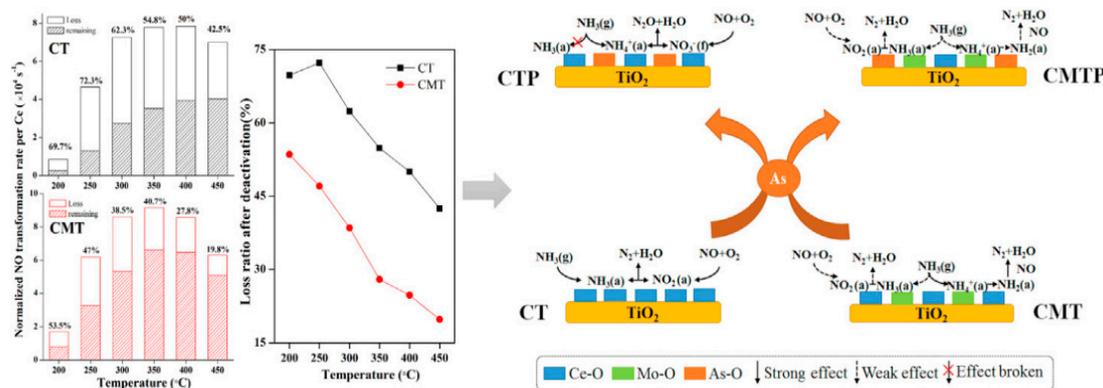


Figure 17. The performance of the arsenic resistance on MoO_3 doped $\text{CeO}_2/\text{TiO}_2$ catalyst for SCR of NO_x with ammonia [101].

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_2O_5 [102], VO_x [103], CuO [104], Nb_2O_5 [105], ZrO_2 [106] and CoO [107]. They can simultaneously enhance the surface acidity, redox performance and $\text{SO}_2/\text{H}_2\text{O}$ 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_2O_5 improved the interaction between CeO_2 and SnO_2 and the redox ability of the catalysts. Then, a novel $\text{V}_2\text{O}_5/\text{CeTiO}_x$ 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_2O . 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 $\text{Nb}_5\text{-Ce}_{40}/\text{Ti}_{10}$ 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_2O_5 can improve the $\text{NH}_3\text{-SCR}$ activity. However, the presence of Nb_2O_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 $\text{NH}_3\text{-SCR}$ 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.

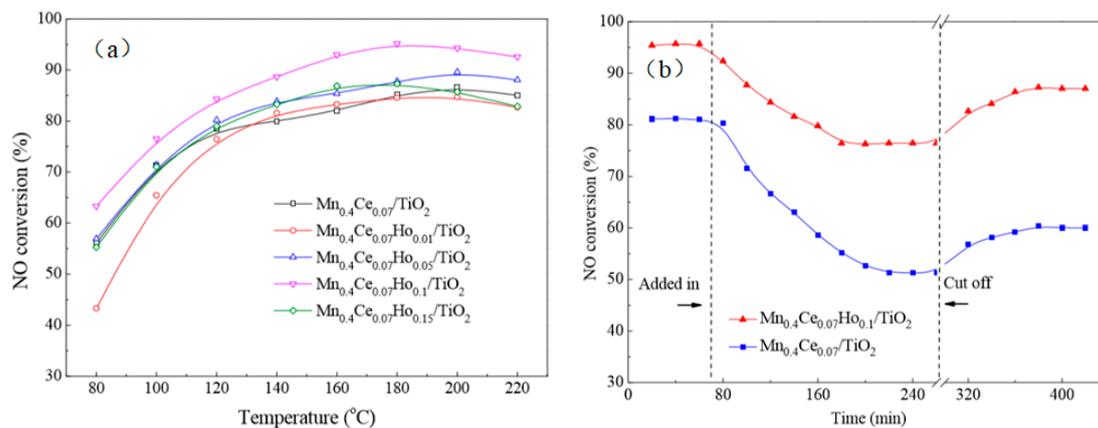


Figure 18. (a) Comparison of catalytic performance of different catalysts and (b) effect of $\text{SO}_2 + \text{H}_2\text{O}$ on NO conversion over $\text{Mn}_{0.4}\text{Ce}_{0.07}/\text{TiO}_2$ and $\text{Mn}_{0.4}\text{Ce}_{0.07}\text{Ho}_{0.1}/\text{TiO}_2$ catalysts [108].

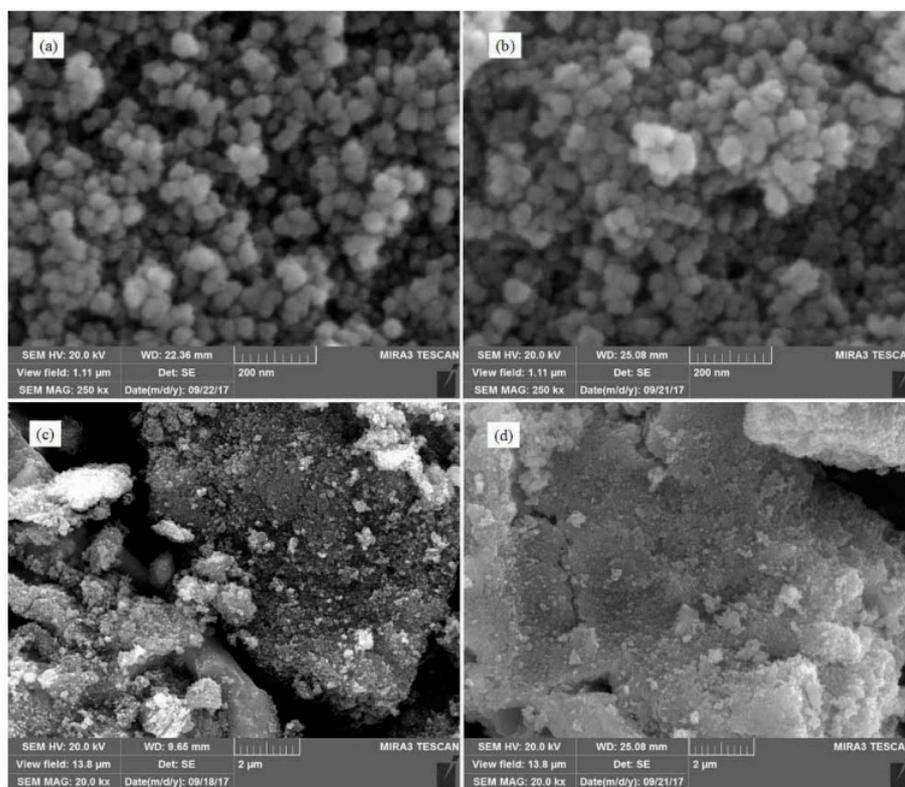


Figure 19. SEM images of two catalysts (a) $\text{Mn}_{0.4}\text{Ce}_{0.07}\text{Ho}_{0.1}/\text{TiO}_2$, (b) $\text{Mn}_{0.4}\text{Ce}_{0.07}/\text{TiO}_2$, (c) $\text{Mn}_{0.4}\text{Ce}_{0.07}\text{Ho}_{0.1}/\text{TiO}_2$ and (d) $\text{Mn}_{0.4}\text{Ce}_{0.07}/\text{TiO}_2$ [108].

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_2O 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 $\text{CuCe}_{0.75}\text{Zr}_{0.25}\text{O}_y/\text{ZSM-5}$ catalyst. The reaction temperature window was widened to 175–468 °C. According to XRD and TEM results, the

presence of Zr increased the dispersion of Cu and inhibited the crystallization of Cu, and XPS and H₂-TPR analysis demonstrated that Cu ions entered the lattice of ZrO₂ or CeO₂.

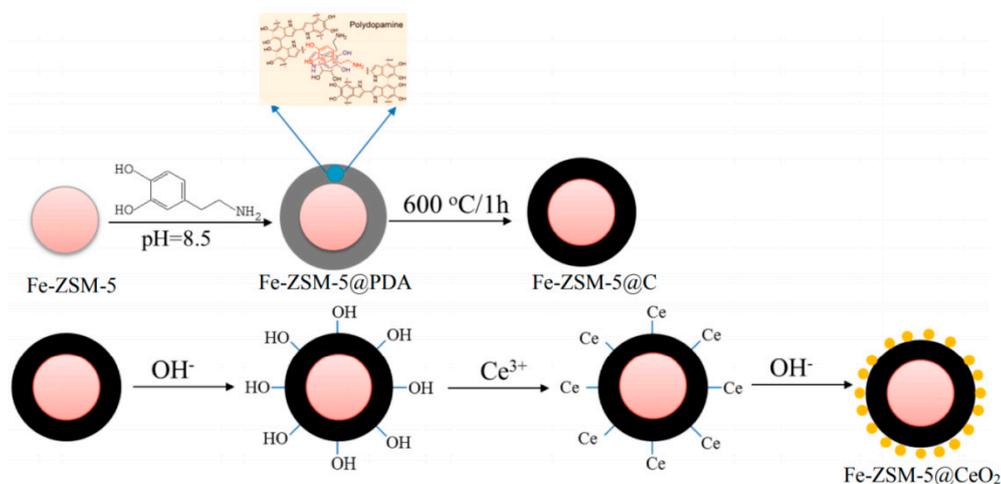


Figure 20. The PDA (Personal Digital Assistant)-assisted route for Fe-ZSM-5@CeO₂ preparation [118].

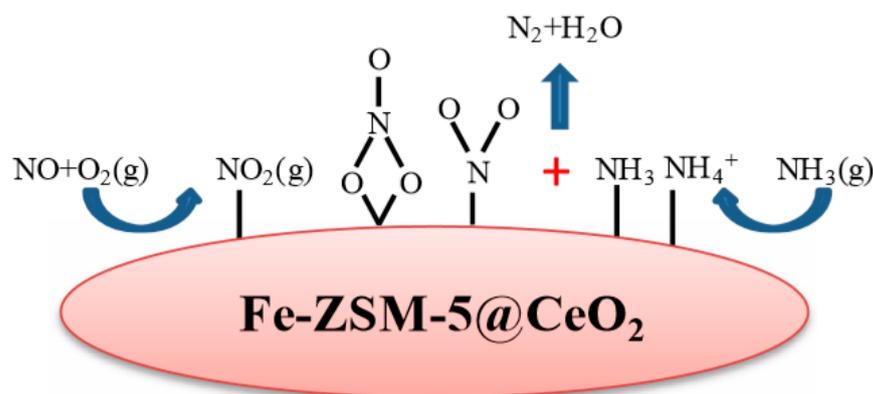


Figure 21. The proposed mechanism of NH₃-SCR over Fe-ZSM-5@CeO₂ [118].

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-NO_x 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% NO_x conversion at 107 °C. The MnO_x 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.

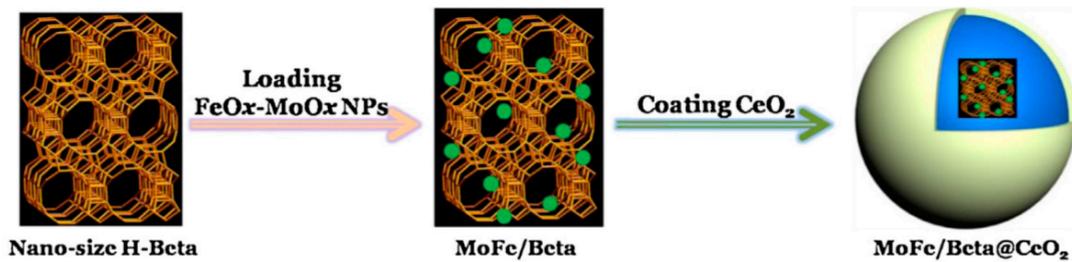


Figure 22. Schematic illustration of the formation of MoFe/Beta@CeO₂ core-shell catalyst [121].

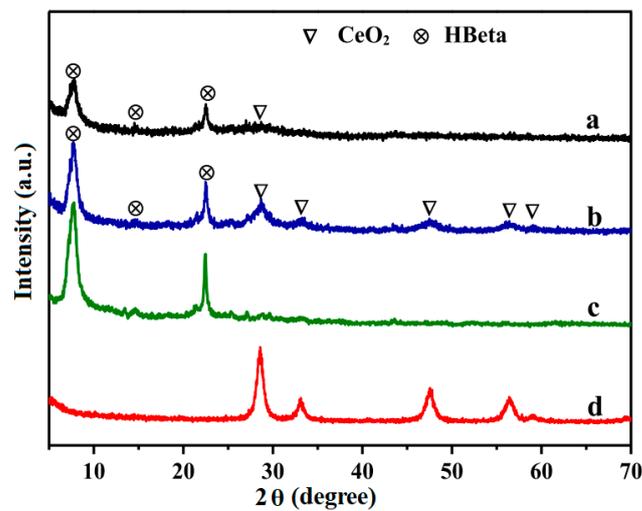


Figure 23. XRD patterns of the catalysts: (a) MoFe/Beta@CeO₂, (b) CeMoFe/Beta, (c) MoFe/Beta and (d) CeO₂ [121].

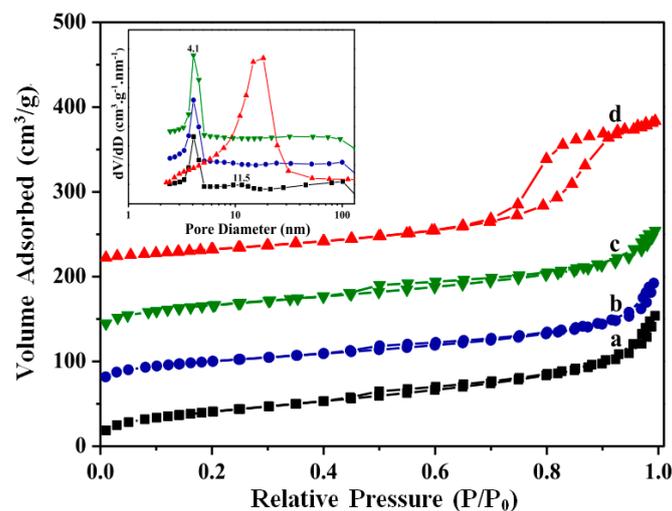


Figure 24. (A) Nitrogen adsorption–desorption isotherms and (B) the size distribution curves of the catalysts: (a) MoFe/Beta@CeO₂, (b) CeMoFe/Beta, (c) MoFe/Beta and (d) CeO₂ [121].

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 SO₂ was simultaneously increased in the catalytic reaction process, thereby resulting in the formation of sulfate on the surface and inhibiting the NH₃-SCR activity of the catalysts. Therefore, the question

of the resistance to SO_2 , H_2O 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/ $^{\circ}\text{C}$	NO_x Conversion/%	Gas Hourly Space Velocity (GHSV)/ h^{-1}	Refs.
$\text{CeO}_2/\text{TiO}_2$	Sol-gel	300–400	93–98%	$50,000 \text{ h}^{-1}$	[31,32]
$\text{CeO}_2/\text{TiO}_2$	Dry ball milling	180	84.6%	GHSV of $30,000 \text{ h}^{-1}$	[34]
$\text{MnO}_x\text{-CeO}_2$	One-step hydrolysis process	180	Over 90%	GHSV of $30,000 \text{ h}^{-1}$	[53]
$\text{CeO}_2\text{-WO}_3$	Two-step hydrothermal impregnation	300–450	100%	GHSV of $60,000 \text{ h}^{-1}$	[58]
$\text{CeO}_2\text{-SnO}_2$	Hydrothermal	280–425	Over 90%	GHSV of $128,000 \text{ h}^{-1}$	[60]
$\text{Mn-Ce}/\text{TiO}_2$	Hydrothermal	150–350	Over 90%	GHSV of $64,000 \text{ h}^{-1}$	[70]
$\text{Ce-W}/\text{TiO}_2$	Sol-gel precipitation	210–460	Over 90%	GHSV of $150,000 \text{ h}^{-1}$	[81]
$\text{Ce-Mo}/\text{TiO}_2$	Sol-gel	250–475	Over 90%	GHSV of $90,000 \text{ h}^{-1}$	[94]
$\text{MnCeW}/\text{TiO}_2$	Impregnation	140–340	Over 95%	GHSV of $40,000 \text{ h}^{-1}$	[83]
$\text{Ce-Cu}/\text{ZSM-5}$	Wet impregnation	210–320	Over 90%	GHSV of $100,000 \text{ h}^{-1}$	[112]
$\text{MoFe}/\text{Beta@CeO}_2$	Wet impregnation	225–600	Over 90%	GHSV of $50,000 \text{ h}^{-1}$	[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_2O and SO_2 . The current study indicated the better catalytic performance of cerium-based bimetallic oxides than pure CeO_2 in $\text{NH}_3\text{-SCR}$. Furthermore, the multiplex oxide catalysts present a wider operation temperature window 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 $\text{NH}_3\text{-SCR}$ 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_2O and SO_2 . Due to that, the improvement of the $\text{SO}_2/\text{H}_2\text{O}$ 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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