A Critical Review of Recent Progress and Perspective in Practical Denitration Application

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Abstract: Nitrogen oxides (NOx) represent one of the main sources of haze and pollution of the atmosphere as well as the causes of photochemical smog and acid rain. Furthermore, it poses a serious threat to human health. With the increasing emission of NOx, it is urgent to control NOx. According to the different mechanisms of NOx removal methods, this paper elaborated on the adsorption method represented by activated carbon adsorption, analyzed the oxidation method represented by Fenton oxidation, discussed the reduction method represented by selective catalytic reduction, and summarized the plasma method represented by plasma-modified catalyst to remove NOx. At the same time, the current research status and existing problems of different NOx removal technologies were revealed and the future development prospects were forecasted.

Keywords: nitrogen oxide; adsorption De-NOx; oxidation De-NOx; selective reduction; nonselective reduction; plasma technology

1. Introduction

NOx is one of the main sources of haze generation and air pollution in all worldwide. On one hand, NOx comes from the natural factors of nitrate caused by decay of organism after biological death and plant straw-burning as well as in the lightning process; on the other hand, it stems from the artificial factors, for instance, iron and steel smelting; petroleum cracking and coal-based fossil fuel power generation; fuel vehicle exhaust emissions [1,2]; and NOx decomposition under the action of soil microorganisms, which is caused by the large amount of nitrogen fertilizer applied in farmland [3,4]. The emission of toxic waste gases has resulted in a significant increase in the content of NOx in the atmosphere, which is damaging to human organs, such as the heart and lungs, and reducing the body’s immunity [5], combining with volatile organic compounds to produce ozone [6], making the human body susceptible to respiratory diseases such as coughs, sore throat, and bronchitis [7,8]; it can also lead to premature birth [9], severely inhibiting photosynthesis, and thus affects plant growth and oxygen conversion [10,11]; but, at the same time, produces photochemical smog [12,13] and acid rain [14]. Furthermore, it forms aerosol with SO2, dust, and water vapor, then produces PM2.5 and other small particle size air pollution suspensions, namely, haze, which cause serious air pollution worldwide [15,16].

Therefore, the treatment of NOx is imminent. The supervision of flue gas emission from fixed source factories and the treatment of motor vehicle exhaust emission from mobile sources have been widely controlled [17] recently, appearing to be the following mature technologies; removal of NOx.
by adsorption, oxidation, reduction, and plasma methods. In view of these attentive findings, adsorption method uses recyclable solid adsorbent material to adsorb NO\textsubscript{x} from flue gas through microporous structure, remarkably, environmentally friendly, no secondary pollution, and energy-saving and -efficient features make it stand out [18–20]. The oxidation process is a method that oxidizes NO into NO\textsubscript{2} or N\textsubscript{2}O\textsubscript{3} with high solubility under the action of an oxidant that is then absorbed by water or alkali solution. The process is simple and cost-saving, and is widely used in the wet flue gas denitrification process with relatively low cost and high removal efficiency of NO\textsubscript{x}; except that the oxidized NO and SO\textsubscript{2} can be removed simultaneously to produce high value-added products [21–23]. The reduction method has the characteristics of high efficiency, cleanliness, and mildness; NO\textsubscript{x} removal can be realized at low temperature at present [24–26]. The plasma method is used to modify the surface of the catalyst, the dispersion of active species can be improved by plasma treatment, and the stability and low temperature activity of catalyst can be improved [27,28].

Seldom, if ever, does a comprehensive article to introduce the current situation and treatment methods of removing NO\textsubscript{x}. In this paper, we tried to renovate, classify, and summarize the significant perspectives and most relevant findings reported in recent research articles and earlier reviews generalizing the mechanism analysis and research progress of NO\textsubscript{x} removal by adsorption, oxidation, reduction, and plasma methods, which can provide means for promoting the technological progress of pollution prevention, maintaining healthy development of factories continuously, studying the methods of removing NO\textsubscript{x} deeply, and mastering different technologies of removing NO\textsubscript{x}, as well as providing guidance for controlling the emission of NO\textsubscript{x} from motor vehicles such as diesel, gasoline, and so on. In order to improve the environmental quality and save the ecological environment, this paper further provides a convenient, low-cost, efficient, and economic guidance line.

2. Removal of NO\textsubscript{x} by Adsorption

The adsorption method for NO\textsubscript{x} treatment is simple to operate, controllable in process, and can capture toxic waste gases with very low concentration efficiently. NO\textsubscript{x} in flue gas is adsorbed by recoverable solid adsorbents through microporous structure [29–31]. The adsorption materials for NO\textsubscript{x} in waste gas mainly include molecular sieve, activated carbon, zeolite, heterophony acid, silica gel, and peat [32]. The adsorption method is efficient and thorough, and can produce nitric acid with high added value. Meanwhile, in recent years, adsorption denitrification technology mainly includes the organic framework of copper (Cu-BTC) adsorption and carbon material adsorption methods [33].

2.1. NO\textsubscript{x} Removed by Cu-BTC Adsorption

Metal–organic frameworks (MOFs) have many advantages, e.g., porous, high specific surface area, and abundant Lewis acidic active sites, and make excellent gas adsorption materials [34], among them, Cu-based MOFs have excellent catalytic performance [35]. Cu-BTC is a special organic metal framework in MOFs. Qin et al. [36] prepared Agx-Cu-BTC dispersed in bimetallic organic framework by preassembly method to absorb NO\textsubscript{x}, this framework had regular octahedral shapes with particle sizes ranging from 10 to 30 microns. Ag-doping into the metal–organic framework of Cu-BTC resulted in a heterojunction, which made the organic functional group, –COO–, blue shift and accompanied with good dispersion of Ag and Cu atoms in the organic framework.

In order to further study the adsorption of NO\textsubscript{x} by Cu-BTC material, Khan et al. [37] studied the adsorption of NO in MOFs-Cu\textsubscript{3}(BTC)\textsubscript{2} and Cu\textsubscript{3}(NH\textsubscript{2}BTC)\textsubscript{2} by \textsuperscript{1}H solid-state NMR, and found that \textsuperscript{1}H in MOFs-Cu\textsubscript{3}(BTC)\textsubscript{2} had a linear relationship with NO adsorption. Kaur et al. [38] explained the synthesis and adsorption properties of Cu-BTC porous octahedral nanocrystals. Meng et al. [39] found that in the framework of Cu-BTC, CO\textsubscript{2} varies with the Langmuir distribution, while the N atom in the NO molecule is mainly responsible for the coordination with the Cu atom. Qin et al. [40] studied the adsorption mechanism of NO on A-Cu-BTC surface (Figure 1a). It was found that the doping of Sr, Ce, Al, and other elements in Cu-BTC could increase the NO conversion (Figure 1b). The modification of Ce by X-ray diffraction analysis enhanced the dispersion of Cu in metal–organic framework BTC (Figure 1c). Scanning electron microscopy further confirmed that the formation of Ce–Cu heterojunction enhanced the low degree (Figure 1d).
2.2. NOx Removed by Carbonaceous Material Adsorption

Removal of NOx by adsorption, especially activated carbon, has undergone a long historical evolution [41]; activated carbon originated from 1900, and its inventor was Raphael von Ostrejko, by whom, British and German patents had been obtained. In 1920, the demand for activated carbon continued to expand through the extensive application of gas masks; at the same time, the application of activated carbon in sewage treatment and catalysis was gradually developed [42,43], activated carbon plants in major industrial countries, including the United States, had been gradually opened and expanded. In the mid-20th century, the field of activated carbon continued to aggrandize and was regarded as “universal adsorbent”. Today, activated carbon is used in pollution control [44,45] with a high-precision instrument to produce pore structure matching the molecular size of the toxic waste gas, which is used to adsorb harmful gases such as benzene, formaldehyde, NOx, NH3, and so on [46,47].

Activated carbon, which is not a natural material, but an artificial material made by processing carbon-containing organic matter, has been widely used as catalyst [48]. Activated carbon comes from a wide range of raw materials, which can be almost any organic compound material, including sawdust, bagasse, branches, shells and other wood materials [49], coking coal, bituminous coal, and other bulk commodities [50]. In the process of activation smelting, a huge specific surface area and complex pore structure are gradually formed, which can effectively remove chroma and odor, and remove most organic pollutants and some inorganic substances in secondary effluent [51], containing some toxic heavy metals [52,53]. Furthermore, activated carbon has excellent thermal and chemical stability, high wear resistance and a variety of oxygen-containing functional groups on the surface, so activated carbon adsorption is a very promising means in the process of flue gas treatment.

Different kinds of oxygen-containing groups are the main active sites on activated carbon [54]; however, the untreated activated carbon has smaller pore volume and specific surface area, higher ash content, and poor adsorption performance. The chemical properties of activated carbon surface
can be changed by chemical oxidation, reduction, and loading [55,56]. Jie et al. [57] found that proper oxidation could improve the degradation ability of activated carbon to nitrogen compounds. Wang et al. [21] found that pretreatment of carrier activated carbon with HNO₃ could improve the NO conversion. Li et al. [58] discovered that the pore size of activated carbon fibers decreased and the surface functional groups increased after modification with Cu₂O/TiO₂, which improved the adsorption capacity of activated carbon fibers for NO. Javier et al. [59] prepared goethite and hematite twin grains on activated carbon, which improved the total denitrification capacity. You et al. [60] caused defects on the surface of activated carbon by changing pore size and nitrogen doping, which greatly improved its NO catalytic oxidation activity at room temperature. Sun et al. [61] used Cu-Fe modified microwave coconut shell activated carbon (CuFe/MCSAC) to remove NO from industrial tail gas, and studied the adsorption process of NO on the surface of activated carbon (Figure 2a). It was found that 3% O₂ was beneficial to increase the conversion of NO, which was attributed to NO oxidation as the first step of selective catalytic reduction reaction (Figure 2b). In addition, the sulfur resistance was studied (Figure 2c,d). The preparation methods and other conditions of different adsorption materials are shown in Table 1.

![Figure 2.](image)

**Figure 2.** (a) Adsorption process of NO on activated carbon surface, (b) effect of adding O₂ on NO conversion, (c) study on SO₂ resistance of Cu-Fe/MCSAC catalyst, and (d) in situ infrared analysis of Cu-Fe/MCSAC after SO₂ entering gas pool [61]. Reproduced from Ref. [61], Copyright 2019, Elsevier.

<table>
<thead>
<tr>
<th>Adsorbent Material</th>
<th>Synthetic Method</th>
<th>Reaction Conditions</th>
<th>Temperature (°C)</th>
<th>Conversion (%)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag-Cu-BTC</td>
<td>preassembled method</td>
<td>500 ppm NO, GHSV 10,000 h⁻¹, N₂</td>
<td>238</td>
<td>100</td>
<td>[36]</td>
</tr>
<tr>
<td>Zeolite</td>
<td>Wetness impregnation</td>
<td>250 mg/m NO, 1000 mg m⁻³ SO₂, GHSV 150 L h⁻¹, N₂</td>
<td>250</td>
<td>91.7</td>
<td>[62]</td>
</tr>
<tr>
<td>Gas-phase</td>
<td>microwave</td>
<td>600 ppm NH₃, 3% O₂, and GHSV 30,000 h⁻¹ N₂</td>
<td>80</td>
<td>50</td>
<td>[61]</td>
</tr>
<tr>
<td>ACF</td>
<td>Precipitation method</td>
<td>Flue gas 0.1–1%, GHSV 1000 mL/min, N₂</td>
<td>40</td>
<td>60</td>
<td>[58]</td>
</tr>
<tr>
<td>AC</td>
<td>Nitric acid</td>
<td>1000 ppm NO, 20 vol.%</td>
<td>25</td>
<td>56.6</td>
<td>[60]</td>
</tr>
</tbody>
</table>
2.3. Other Crucial NOx Adsorbents

There are numerous kinds of NOx adsorbents; low-priced and efficient adsorbents are required for denitrification. Li et al. [63] prepared fly ash derivative Cu/SAPO-34 by acid–alkali combined hydrothermal method to absorb NOx. Mire et al. [64] studied the removal of NOx using H-ZSM-5 as adsorbents, and found that Fe-modified Fe/H-ZSM-5 had the strongest adsorption capacity. Chen et al. [65] synthesized Fe-ZSM-5@CeO2 adsorbent for adsorbing NOx. Baran et al. [66] prepared Cu-BEA molecular sieve to study the adsorption characteristics of NOx. Ma et al. [67] developed a new Ag nano-g-C3N4/WS2 material to adsorb NOx. Imai et al. [68] developed a niobium phosphate adsorbent to remove NOx. Xiao et al. [69] studied the adsorption of NOx on mesoporous molecular sieve doped with platinum (Pt/SBA-15). Wang et al. [70] found that when iron and cobalt were loaded on the active semi-coke material; it was beneficial to NOx adsorption. Chen et al. [71] studied NO removal performance based on sinter adsorption materials. The synthetic methods and reaction conditions of different adsorption materials are shown in Table 2.

<table>
<thead>
<tr>
<th>Adsorbent Material</th>
<th>Synthetic Method</th>
<th>Reaction Conditions</th>
<th>NO Conversion %</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu/SAPO-34</td>
<td>Acid–alkali hydrothermal</td>
<td>300 ppm NO, 3% O2, GHSV 12,000 h⁻¹, N2</td>
<td>90</td>
<td>[63]</td>
</tr>
<tr>
<td>Fe/H-ZSM-5</td>
<td>Hydrothermal</td>
<td>5000 ppm NO, 5% O2, GHSV = 35,000 h⁻¹, He</td>
<td>63.4</td>
<td>[64]</td>
</tr>
<tr>
<td>Fe-ZSM-5@CeO2</td>
<td>Dopamine polymerization</td>
<td>1000 ppm NO, 5% O2, GHSV 33,600 h⁻¹, N2</td>
<td>90</td>
<td>[65]</td>
</tr>
<tr>
<td>Cu-BEA</td>
<td>Precipitation</td>
<td>1000 ppm NO, 3.5 vol% O2, GHSV 1000 mL/min, N2</td>
<td>100</td>
<td>[66]</td>
</tr>
<tr>
<td>g-C3N4/WS2</td>
<td>Solvent evaporation</td>
<td>500 ppm NO, 20vol% O2, GHSV 400 mL/min, N2</td>
<td>72.5</td>
<td>[67]</td>
</tr>
<tr>
<td>Niobium phosphate</td>
<td>Impregnation</td>
<td>1000 ppm NO, 1125 ppm O2, GHSV 100 mL/min, He</td>
<td>100</td>
<td>[68]</td>
</tr>
<tr>
<td>Pt/SBA-15</td>
<td>Thermal hydrolysis</td>
<td>4000 ppm NO, 10 vol% O2, GHSV 50 mL/min, Ar</td>
<td>100</td>
<td>[69]</td>
</tr>
<tr>
<td>Semi-coke</td>
<td>Hydrothermal</td>
<td>1000 ppm NO, GHSV 6000 h⁻¹, N2</td>
<td>100</td>
<td>[70]</td>
</tr>
<tr>
<td>Sintered ore</td>
<td>Impregnation</td>
<td>400 mg/m³ NO, 15% O2, GHSV = 1000 h⁻¹, Ar</td>
<td>61.6</td>
<td>[71]</td>
</tr>
</tbody>
</table>

Chemical adsorption usually requires a certain activation energy, and the adsorption rate is slow at low temperature [72]. The adsorption method is closely related to the surface chemical properties of adsorbents, and it can not only eliminate the pollution of NOx to the atmosphere, but recycle them to produce high value-added products. However, due to its limited adsorption capacity, the size of the absorber is huge, the adsorbent needs to be regenerated frequently, and the one-time investment is large. In particular, activated carbon has the possibility of spontaneous combustion above 300 °C, which causes considerable difficulties in the regeneration of adsorbents and limits its application.

3. Removal of NOx by Oxidation

Since MeKee oxidized low-concentration NO to NO2 in 1921; it laid the foundation for NO oxidation [73]. In the early 1980s, Japanese scholars made a more in-depth study on the oxidative denitrification. At this time, NO oxidative denitrification technology rose gradually. In the late 1990s, Takashi Ibusuki et al. [74] found that TiO2 can oxidize NO to HNO3 under ultraviolet irradiation.
rapidly. Nowadays, the technology of removing NOx by oxidation is relatively mature. It can not only control the morphology of catalysts by some means, but study the law that affects the activity of catalysts. At the same time, investigators often only study the activity of catalysts in a low-temperature window gradually [75–77]. This method uses oxidizer to oxidize NO, which is not soluble in water, to NO2 or N2O3 in flue gas [78–80], which is then sprayed and washed by water, alkali solution, acid solution, or metal complex solution so that the NOx in the gas phase are transferred to the liquid phase to realize the denitrification treatment of the flue gas. O3, ClO2, NaClO2, NaClO, KMnO4, H2O2, Cl2, and HNO3 can be used as oxidants [81]. According to the types of oxidation technology, it can be divided into gas-phase oxidation, liquid-phase oxidation, and catalytic oxidation.

3.1. NOx Removed by Gas-phase oxidation

Since the development of a low temperature oxidation technology called LoTOx by Linde Industrial Gases Company in the United States, more and more researchers have been studying the denitrification performance of O3. The gas-phase oxidation method has been used to purify flue gas from boilers fueled with natural gas, and the denitrification rate is over 90%. The purification process is simple and has no effect on the normal operation of the boiler. It can recover high-grade HNO3 [82,83]. O3 is highly oxidizable, and the reduction potential in the process of O3 ↔ O2 can reach 2.07 eV, which makes the oxidation of NO possible.

At present, ClO2 and O3 are widely used in oxidative denitrification as gas phase oxidants. Hultén et al. [84] found that when the molar ratio of ClO2 to NO was 0.6 at 160 °C, the total amount of NOx removed was 94%. Sun et al. [85] discovered that the removal efficiency of NO increased with increasing O3 concentration. The removal efficiency of NOx reached 82% when the flue gas temperature exceeded 40 °C. Mok et al. [86] proposed a two-step process consisting of an O3 chamber and an absorber containing a reductant solution for simultaneous removal of NOx. When the flue gas passes through the O3 chamber, and in turn the absorber, the removal rate of NOx is ~95%. Lin et al. [87] studied the deep oxidation of NO by O3 on the MnOx-based catalyst. The experimental device of deep oxidation showed that the catalyst had good stability and sulfur resistance. Han et al. [88] prepared the black TiO2 catalyst by sol–gel method. The deep oxidation of NO to HNO3 was further strengthened (Figure 3a): black TiO2 had higher NO conversion than TiO2 at the low temperature of 60 °C (Figure 3b). New peaks of black TiO2 at 532.6 eV were found by X-ray photoelectron spectroscopy (XPS) chromatography, which further confirmed the excellent catalytic activity of black TiO2 (Figure 3c). In addition, the sulfur resistance of black TiO2 was investigated (Figure 3d).
3.2. NO\textsubscript{x} Removed by Liquid-Phase Oxidation

The liquid-phase oxidation absorption method is that NO is oxidized by liquid phase oxidant and then absorbed by alkali absorption method. The de-NO\textsubscript{x} rate can reach 90–95% when using KMnO\textsubscript{4} or NaClO\textsubscript{2} as liquid phase oxidizers [89]. NaClO\textsubscript{2} has strong oxidizing property; its solution can oxidize NO to NO\textsubscript{2}, and then NaNO\textsubscript{3} was obtained. It is easy to oxidize NO with NaClO\textsubscript{2} and the removal rate of NO is high [90]. In order to cope with the increasingly severe environmental problems and find effective ways to control air pollution, the NO gas discharged from flue gas can be removed simultaneously through a wet NaClO\textsubscript{2} scrubber combined with a plasma electrostatic precipitator.

Deshwal et al. [91] used NaClO\textsubscript{2} solution to remove NO\textsubscript{x} from simulated flue gas. NaClO\textsubscript{2} was a better oxidant when the pH was less than 4. NaClO\textsubscript{2} was decomposed into ClO\textsubscript{2} gas in acidic medium. It was considered that NaClO\textsubscript{2} participates not only in NO oxidation but in NO\textsubscript{2} absorption. Hao et al. [92] prepared a new composite oxidant NaClO\textsubscript{2}/Na\textsubscript{2}S\textsubscript{2}O\textsubscript{8}, removing SO\textsubscript{2} and NO simultaneously by pre-oxidation. The removal rates of SO\textsubscript{2} and NO were 100% and 82.7%, respectively, under the optimum conditions. Yang et al. [93] used ultraviolet radiation sodium chlorite (UV/NaClO\textsubscript{2}) solution to remove NO\textsubscript{x} from simulated flue gas in a small scrubbing reactor. The results showed that the addition of Cu and Mn resulted in the lattice expansion and micro-strain decrease of CeO\textsubscript{2}-ZrO\textsubscript{2}, which led to the formation of oxygen vacancies. Besides, there was a strong interaction between surface Cu, Mn, and Ce through charge transfer.

3.3. NO\textsubscript{x} Removed by Catalytic Oxidation

O\textsubscript{3} has a high preparation cost in gas-phase oxidation, which leads to its narrow promotion area; the liquid-phase oxidation method requires higher corrosion resistance of equipment, and the price of oxidant is relatively expensive, which restricts its industrial application. Therefore, the preponderance of catalytic oxidation method are beginning to highlight [94]. The oxidant is the excess oxygen in flue gas, the catalysts are vanadium, tungsten, and titanium and rare earth metal oxides supported on activated carbon, alumina, and silica. This method has been widely used in flue gas
purification with the denitrification rate is over 90% [95]. The preponderance of catalytic oxidation are closed-circuit circulation; simultaneous removal of SO$_2$, dust, and other pollutants without additional oxidizer; injection of 5% steam into flue gas can improve the efficiency and life of catalyst; and low investment and operation cost of equipment. Catalytic oxidation technology for NO$_x$ removal includes molecular sieve catalytic oxidation, metal oxide catalytic oxidation and Fenton system catalytic oxidation, which have their respective applicable conditions.

3.3.1. Catalytic Oxidation of Molecular Sieve

A kind of natural aluminosilicate, which has the functions of ion exchange, molecular sieving, catalysis and adsorption, called zeolite, and synthetic zeolite is also called molecular sieve. The basic structure of molecular sieve skeleton is Al$_2$O$_3$ and SiO$_4$ tetrahedrons, which form a three-dimensional network structure by combining the common O atoms [96]. It has strong selectivity, high-temperature resistance, and high adsorption capacity, which is widely used in organic chemical industry and petrochemical industry. It is also an excellent adsorbent for gas dehydration. At the same time, more and more attention has been paid to the purification of waste gas. Molecular sieves have regular and uniform intracrystalline channels, and the pore size is close to the molecular size. The catalytic performance of molecular sieves varies significantly with the geometrical size of reactant molecules, product molecules or reaction intermediates [97].

Molecular sieve catalysts for NO treatment are mostly used in oxidation and decomposition processes, such as octahedral molecular sieve (OMS-2) catalysts. Molecular sieve catalysts for oxidation reaction are mainly transition metal ion exchange zeolites and specific types of zeolite zeolites, and these zeolite catalysts only show high temperature activity. Liu et al. [98] first used solvent-free synthesis of cryptomelane OMS-2 to oxidize waste material in coal-fired flue gas, which provided a new idea for oxidative denitrification by molecular sieve. Geng et al. [99] prepared zinc oxide nanoparticle molecular sieves by melt infiltration method to remove exhaust gas at room temperature. Saminda et al. [100] synthesized Mn octahedral molecular sieve (OMS-2) to remove flue gas by catalytic oxidation. Park et al. [101] studied the equilibrium and kinetics of adsorption of N$_2$O on carbon molecular sieves, concluding that the adsorption rate of N$_2$O was influenced by kinetics and Lewis structure. Sarmah et al. [102] analyzed the microstructure and morphology of OMS by scanning electron microscopy, which showed the highly cross-linked nanowire morphology. Hamaguchi et al. [103] studied the mechanism of NO removal on the surface of molecular sieve K$_{1.5}$Mn$_4$O$_{10}$ (Figure 4a), finding that the thermal unstable oxygen potential acted as the oxidation site of NO at low temperature. The doping of K increased the adsorption of NO (Figure 4b). The particle size of K was 0.46 nanometers (Figure 4c). NO generated in K-OMS-2 molecular sieves NO$_3^-$, the formation of NO$_3^-$ is proved by Kubelka–Mung theory (Figure 4d).
3.3.2. Catalytic Oxidation of Metal Oxides

Metal oxide, the compound of O and another metal element, is a kind of excellent catalyst. As the main catalyst, promoter and carrier, it has been widely used in the field of catalysis. After nanotreatment of metal oxides, nano-materials with uniform dispersion, small particle size, and high purity can be obtained, and their catalytic performance is better [104]. Metal oxides provide oxygen free radical formation products for reactants and provide active sites for adsorption (or coordination) of reactants to make them active as well.

Metal oxide is a kind of catalyst that has been studied earlier in the field of NO catalytic oxidation and developed vigorously. Jogi et al. [105] found that the presence of TiO₂ greatly enhanced the efficiency of O₃ oxidation of NO₂ to N₂O₅ at 100 °C. Wang et al. [106] synthesized manganese oxides with different valences (MnO₂, Mn₂O₃, and Mn₃O₄) by hydrothermal method. At the space velocity of 48,000 mLg⁻¹h⁻¹, with oxygen as oxidant, the maximum NO conversion rate was 91.4%. Ma et al. [107] synthesized nanoparticle-like monocrystalline SmMn₃O₈ by adjustable hydrothermal method; among them, nanoparticle-like SmMn₃O₈ achieved more than 90% NO conversion at 300 °C. Jia et al. [79] synthesized a series of Mn–Fe catalysts for oxidative denitrification by hydrothermal method. It was found that the addition of iron oxides could effectively improve the catalytic oxidation performance of manganese dioxide at low temperature. Cai et al. [108] synthesized Cr–Ce oxidative denitrification catalyst with three-dimensional structure by hydrothermal method; more active sites could be exposed with the increase of surface particle size and roughness (Figure 5a). The chromium-Ce/Et catalyst synthesized with ethanol as solvent had the highest catalytic oxidative denitrification activity (Figure 5b). Through a series of characterization catalysts, the Cr–Ce/Et coexistence effect formed the phase interface between atoms (Figure 5c). In addition, the effects of steam and SO₂ on NO conversion over Cr–Ce/Et catalysts at 250 and 300 °C were studied (Figure 5d).
3.3.3. Photocatalytic Oxidation

Photocatalytic oxidation is a new process of NOx treatment, which has been developed gradually over the last ten years. The principle of photocatalytic oxidation is to irradiate semiconductor catalysts with light of specific wavelength, to stimulate valence band electrons on semiconductor materials to transit into conduction band, and to generate holes in valence band. Conduction band electrons and valence band holes have strong reductive and oxidative properties, respectively. When they are in contact with flue gas, H2O, O2, and NOx adsorbed on the catalyst surface will generate active free radicals under the action of light, and then catalytic oxidation reaction will take place to oxidize NOx to NO3−. Because it does not need to inject additional reductant and is simple to operate with low-cost and no secondary pollution, it is the focus of photocatalytic technology research at present [109,110].

The catalysts for photocatalytic oxidation are mainly metal oxide semiconductor materials. Among them, TiO2 has the merits of high catalytic activity, photochemical stability and low price. It is the most commonly used catalytic material in photocatalytic reaction. Duan et al. [111] found that a small amount of Ag nanoclusters supported on TiO2 was beneficial to improve photocatalytic activity. He et al. [112] synthesized carbon-encapsulated nanocrystalline TiO2 catalyst, which achieved 71% NO conversion under visible light. Yuan et al. [113] prepared TiO2 aluminosilicate fiber nanocatalyst for photocatalytic removal of NOx. Jin et al. [114] prepared SrTiO3 catalyst with SrCO3 successfully as auxiliary by one-step in situ pyrolysis for removal of NOx. Huy et al. [115] reported for the first time the removal of NOx by visible light on SnO2/TiO2 nanotube heterojunction catalysts. The reaction mechanism was shown in Figure 6a. The removal efficiency of NOx under visible light was represented in Figure 6b. Figure 6c shows that SnO2/TiO2 nanotube heterojunctions had been successfully synthesized. Figure 6d proves that the superoxide anion radical played an important role in visible light photocatalytic oxidation by using active oxygen detection materials.
Figure 6. (a) Reaction mechanism of visible light removal of NOx by SnO2/TiO2 nanotube heterojunction, (b) removal efficiency of NOx under different conditions, (c) transmission electron microscopy of SnO2/TiO2 nanomaterials, and (d) detection process of free radicals under different conditions [115]. Reproduced from Ref. [115], Copyright 2019, Elsevier.

3.3.4. Catalytic Oxidation of Fenton System

The essence of Fenton system is that the chain reaction between Fe2+ and H2O2 catalyzes the production of OH· with strong catalytic oxidation ability. Its oxidation potential is only 2.80 V after that of fluorine, which is a strong oxidation system. In addition, OH· has high electrophilicity and electronegativity and its electron affinity can reach 569.3 kJ, which is associated with strong additive reaction properties. Therefore, Fenton reagent can selectively degrade most organic compounds in water, especially suitable for the treatment of organic wastewater which is difficult to be degraded by biology or chemical oxidation. The prospect of catalytic oxidation of NOx in flue gas is also extremely promising [116,117].

NO can be oxidized efficiently at low temperature with H2O2. At present, Fenton reagents and Fenton-like reagents are widely studied [118,119]. H2O2, no secondary pollution and low price, is one of the main components of Fenton reagent and can achieve high denitrification rate [120]. Fenton reagent refers to the H2O2/Fe2+ system, through a series of catalytic actions of Fe2+ to produce iso-strong oxides OH·, HO2·, and O2-. Wang et al. [121] adopted a new Fenton process catalyzed by Cu2+ and Fe2+ in coordination, which inducing more OH· to deoxidize NO, and enhancing the denitrification activity of Fenton catalyst. Zhao et al. [122] carried out simultaneous denitrification of flue gas in bubbling reactor with Fenton reagent. The removal rate of NO was over 90%. Cheng et al. [123] found that too high Fe2+ concentration and pH were not conducive to NO oxidation and when the pH was only 2–5, the formation rate of OH reached the maximum. Guo et al. [124] confirmed that the oxidation of NO by Fenton reagent occurred on the liquid membrane side, low concentration of NO was not conducive to mass transfer to the liquid side. Cui et al. [125] took fly ash as raw material and removed NO with hydrogen peroxide. It is found that Fenton heterogeneous catalyst modified by alkali, acid, and alkali has high catalytic activity. Zhao et al. [126] used heterogeneous ultraviolet spectrophotometry to remove SO2 and NO from flue gas for the first time. Yuan et al. [127] oxidized NO with H2O2 as oxidant in the two-ion Fenton reaction system (Figure 7a,b). The modification of Ce could significantly improve the conversion of NO (Figure 7c). The effect of Ce on the dispersion of Fe2+ was further investigated by XRD (Figure 7d).
In recent years, oxidation method has been widely used in the United States and European countries to remove NOx. Selective catalytic oxidation method has the advantages of relatively low cost and high removal efficiency of NOx. The oxidized NO can be removed simultaneously with SO2 to produce high value-added products. The process is simple and cost-saving [128,129]. The reagents materials and other conditions of different oxidation methods are shown in Table 3.
Table 3. Removal of NOx by oxidation.

<table>
<thead>
<tr>
<th>Oxidation Method</th>
<th>Reagents Materials</th>
<th>Temperature (°C)</th>
<th>NO Conversion (%)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas phase</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>ClO2</td>
<td>160</td>
<td>94</td>
<td></td>
<td>[84]</td>
</tr>
<tr>
<td>O3</td>
<td>40</td>
<td>82</td>
<td></td>
<td>[85]</td>
</tr>
<tr>
<td>O3</td>
<td>60</td>
<td>95</td>
<td></td>
<td>[88]</td>
</tr>
<tr>
<td>Liquid phase</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>UV/NaClO2</td>
<td>50</td>
<td>98.1</td>
<td></td>
<td>[92]</td>
</tr>
<tr>
<td>NaClO/NaClO2</td>
<td>50</td>
<td>85</td>
<td></td>
<td>[90]</td>
</tr>
<tr>
<td>NaClO2/Na2S2O8</td>
<td>120</td>
<td>82.7</td>
<td></td>
<td>[92]</td>
</tr>
<tr>
<td>Catalytic oxidation</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>K-OMS-2</td>
<td>50</td>
<td>-</td>
<td></td>
<td>[103]</td>
</tr>
<tr>
<td>MnO2</td>
<td>250</td>
<td>91.4</td>
<td></td>
<td>[106]</td>
</tr>
<tr>
<td>SmMn2O5</td>
<td>300</td>
<td>90</td>
<td></td>
<td>[-107]</td>
</tr>
<tr>
<td>Fe0.32MnO2</td>
<td>250</td>
<td>80</td>
<td></td>
<td>[79]</td>
</tr>
<tr>
<td>Fenton</td>
<td>140</td>
<td>90</td>
<td></td>
<td>[125]</td>
</tr>
<tr>
<td>La0.8Pr0.2MnO3</td>
<td>260</td>
<td>91</td>
<td></td>
<td>[130]</td>
</tr>
</tbody>
</table>

4. Removal of NOx by Reduction

As early as the late 1970s, reduction denitrification technology was put into use in Japan. Over time, reduction technology had been widely used in Europe and the United States, and is considered to be the most advantageous technology for removing NOx. Because of the limitations of NOx control before and during combustion, the control of NOx in flue gas after combustion has attracted wide attention [131]. There are many technologies for removing NOx from flue gas, according to different principles, they can be divided into catalytic reduction and adsorption, besides, in the view of different working media, and they can be divided into dry and wet methods. Different treatment methods are selected according to the concentration of NOx tail gas and other working conditions [132–134]. Reduction denitrification technology includes selective noncatalytic reduction and selective catalytic reduction.

4.1. Selective Noncatalytic Reduction (SNCR) for NOx Removal

Selective noncatalytic reduction (SNCR) has been developed earlier and its technology is relatively mature, which can generally reduce NOx by 50 to 60%. The main principle is that under the condition of catalyst, ammonia, or urea with reductive groups are injected into the flue gas of high temperature furnace was between 850 and 1100 °C. NOx in the flue gas is reduced to harmless nitrogen after atomizing and spraying into the reactor. The advantage of SNCR technology is that there is less investment and occupied area a relatively small, SNCR technology has certain limitations and is generally only suitable for small coal-fired boilers with low NOx content [135]. Since there is no catalyst to accelerate the reaction, the operating temperature is higher than that of SCR method. In order to avoid NH3 being oxidized, the temperature should not be too high. The current trend is to use urea instead of NH3 as reducing agent.

In order to further study the development status of selective noncatalytic reduction (SNCR), Hao et al. [136] found that in the process of SNCR containing NH3, the use of water vapor could increase NO reduction rate, and the optimum water vapor content was 4–8%. Sodium and potassium additives could promote NO reduction in the order of Na2CO3 > KCl > NaCl. Fu et al. [137] found that CaO had an effect on the pyrolysis of urea during melt coalescence, thus reduced the pyrolysis rate of urea and the performance of urea SNCR denitrification process. Chen et al. [138] found that when urea was added into hydrazine hydrate solution, better denitrification efficiency could be obtained in the temperature range of 550 to 650 °C. Kang et al. [139] validated the optimized SNCR reaction mechanism, and combined the optimized mechanism with CFD software to simulate the SNCR denitrification process. Fu et al. [140,141] studied the reaction mechanism of SNCR denitrification on CaO and CaCO3 surfaces (Figure 8a). NH3 conversion and NO selectivity in different mixed gas without oxygen were analyzed (Figure 8b), HCHO was the main product of NH3 transformation on
CaCO₃ surface (Figure 8c), and in situ infrared spectroscopy further confirmed the species of reaction intermediates (Figure 8d).

![Figure 8](image)

**Figure 8.** (a) Reaction mechanism of selective noncatalytic reduction (SNCR) on CaCO₃ surface, (b) NH₃ conversion and NO selectivity in different mixed gas, (c) FTIR analysis of CaCO₃ surface active species, and (d) in situ FTIR analysis of reaction intermediates at 500 °C [140,141]. Reproduced from Ref. [140], Copyright 2016, Elsevier; Reproduced from Ref. [141], Copyright 2015, Elsevier.

### 4.2. Selective Catalytic Reduction (SCR) for NOₓ Removal

SCR is one of the most widely used denitrification methods at home and abroad at present. It was first developed by the United States, and then widely used in industrial production in Japan. SCR can make the removal efficiency of NOₓ reach more than 90% [142,143]. The basic principle of SCR technology is that under the existence of catalyst, NH₃, CO, and other reducing agents are used to reduce the NOₓ to N₂ in flue gas. At the same time, this method also has some shortcomings. The core of SCR technology lies in the selection of excellent catalysts. The complex composition of flue gas can easily deactivate and poison the catalysts. Especially for water and sulfur, SCR catalysts are more sensitive, which affects the life of the catalysts, and makes the cost of treatment and operation significantly higher [144–146]. SCR technology is a method of selectively reducing NOₓ to nitrogen and water by using ammonia or hydrocarbon or carbon monoxide as reductant under certain temperature and catalyst [147,148]. SCR is the most mature and widely used denitrification technology with high denitrification efficiency. It is the most efficient means to control NOₓ pollution. Generally, the denitrification efficiency can reach 80–90% based on the rational selection of reactor and catalyst [149].

#### 4.2.1. NH₃-SCR

NH₃-SCR is one of most efficient processes for denitrification. Up to date, NH₃-SCR systems have been widely applied in stationary industrial installations (SIIs), such as power plants and industrial boilers. Commercial vanadium-based catalysts usually work in the temperature range of 300 to 400 °C, and often suffer from high dust and SO₂ contents in flue gases, while a manganese-based monolithic honeycomb catalyst (MHC) showed high activity in NH₃-SCR at room temperature (as
low as 20 °C). We believe that this room temperature NH₃-SCR technology is effective for high concentration NOₓ removal with great potential for applications in SIIs.

Current SCR denitration (De-NOₓ) technologies come in three categories, high-temperature De-NOₓ at high dust content, high-temperature De-NOₓ at high SO₂ content, and low-temperature De-NOₓ at high H₂O content, as shown in Figure 9. These applications are particularly challenging because of the varying operation conditions of SIIs with regard to flue gas temperature. The general niches and limitations of monolithic honeycomb catalysts (MHCs) for SCR processes and practical NOₓ removal can be highlighted as follows.

(i) High-temperature De-NOₓ unit at high dust content. In this unit, the temperature of the flue gas entering the NH₃-SCR reactor is as high as 300–400 °C. This high temperature significantly favors the De-NOₓ performance of most of the catalysts (e.g., vanadium-type-based catalysts) [150,151]. At present, this technology is the most widely used in SIIs. However, further progress in promising applications is still somewhat bottlenecked by (a) the high dust content in the flue gas, which easily leads to erosion and plugging holes in MHCs and (b) the high SO₂ content of the flue gas, leading to catalyst poisoning and deactivation. These limitations are yet to be overcome [152].

(ii) High-temperature De-NOₓ unit at high SO₂ content. In this unit, the temperature of the flue gas entering the NH₃-SCR reactor ranges from 180 to 280 °C. A dust-removing apparatus is used to mitigate the erosion and plugging issues for longer service life of MHCs for NH₃-SCR. Nevertheless, the high SO₂ content continues to raise greater worries for activity declining and shorter service life [153,154]. Most of manganese- and copper-based catalysts with high SO₂ resistance could be used in this process [155,156].

(iii) Low-temperature De-NOₓ unit at high H₂O content. In this unit, the temperature of the flue gas entering the NH₃-SCR reactor is below 160 °C. Although dust and SO₂ are previously removed, the H₂O content in the flue gas still remains as high as 10 vol.%. Almost all current catalysts show low activity and poor water-resistance at this temperature [157,158]. Therefore, this technique is not practical for SIIs.
Figure 9. (a) Design route for De-NOx via NH3-SCR. (I) high-temperature at high dust content, (II) high-temperature at high SO2 content, (III) low-temperature at high H2O content, and (IV) low temperature with water removal. (b) Room temperature De-NOx via NH3-SCR. (c) Luo’s process for treating CO, NOx, and SOx [159]. Reproduced from Ref. [159], Copyright 2018, Elsevier.

Additionally, in the process of low-temperature De-NOx unit after H2O removal, the water vapor in the flue gas is condensed and recovered. Furthermore, the flue gas obtained after the dust removal, desulfurization, and water recovery steps can be heated to a temperature ranging from 50 to 150 °C, depending on the flue gas flow. Most Mn-based catalysts exhibit great NH3-SCR performance at low temperature [160]. This technique has potential in application for SIIs; however, the NO removal at low temperature still exists as a big challenge.

Zhang et al. [161] found inspiring room temperature De-NOx performance in NH3-SCR with Mn-based MHCs. The MnOx-Fe2O3/vermiculite (VMT) catalyst showed high N2 selectivity of 97.1% at 20 °C. The corresponding MnOx-Fe2O3/VMT MHC exhibited a NO conversion of 62.2% at NO content of 500 ppm. Additionally, MnOx-Fe2O3/VMT MHC showed excellent SO2 resistance even at SO2 content of 300 ppm, at which sulfate might be easily formed to foil the catalyst surface. As shown in Figure 8b, this room temperature NH3-SCR technology is effective for high concentration NOx removal, showing great potential for applications in SIIs.

For low temperature NH3-SCR denitrification catalyst, it is very crucial to study denitrification mechanism for further improving its activity [162,163]. Ryu et al. [164] studied the location of active sites in NH3-SCR reaction, mainly at Lewis acid sites on the catalyst surface. Campisi et al. [165] functionalized HAP by ion exchange. The high dispersion of Fe3+ species ensured the acid and redox centers. Yang et al. [166] studied the mechanism of NH3-SCR reaction of NO on CuMnO4 catalyst. Density functional theory calculation showed that the dehydrogenation of NH3 was the decisive step of N2 formation. Wu et al. [167] used hydrotalcite-like precursor to induce the modification of NiO by titanium dioxide (Figure 10a), which produced a good interface effect in the calcination process.
The active components were dispersed at atomic level, and the conversion rate was over 90% in the temperature range of 240 to 360 °C (Figure 10c,d).

Figure 10. (a) Different preparation processes of Ni–Ti hydrotalcite-like precursors, (b) NO conversion of catalysts obtained by different preparation processes, (c) mechanism of NO removal induced by TiO$_2$ on the surface of NiO modified catalysts, and (d) sulfur resistance of Ni–Ti hydrotalcite-like precursors at 240 °C [167]. Reproduced from Ref. [167], Copyright 2019, Elsevier.

MnO$_x$-based catalysts are currently the focus of low-temperature NH$_3$-SCR flue gas denitrification technology research [168–170]. As an active component, MnO$_x$ can provide free electrons, which play an important role in the SCR process [171]. Wang et al. [172] prepared MnO$_x$ catalyst with Na$_2$CO$_3$ as precipitator has a high specific surface area and amorphous framework structure. Zhang et al. [173] synthesized Fe modified MnO$_x$/TiO$_2$ catalyst by sol–gel method; when the flue gas was simulated at 250 °C, the NO conversion reached 76.8%. Wang et al. [174] developed a MnO$_x$–CeO$_2$–Al$_2$O$_3$ catalyst with enhanced activity and good water resistance by using instantaneous nano-precipitation technology. Xiang et al. [175] discussed the adsorption mechanism of NH$_3$ and NO on MnO$_x$-based catalysts supported on γ-Al$_2$O$_3$. Wang et al. [176] studied the performance of a series of W-modified MnO$_x$/TiO$_2$ catalysts for NH$_3$-SCR. In situ infrared spectroscopy showed that more Brønsted and Lewis acid sites were formed on the catalyst surface after tungsten doping. Zhang et al. [177] studied the effect of Ce-Mo modified TiO$_2$ adsorbent on NO adsorption. Zhao et al. [178] prepared two-dimensional layered MnAl double oxides by instantaneous nanoprecipitation. Wang et al. [179] prepared MnO$_x$–CeO$_2$–Al$_2$O$_3$ catalysts with different MnO$_x$ content by self-propagating high-temperature synthesis firstly, and the effects of different synthesis methods on NO conversion were compared in Table 4.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Synthetic Method</th>
<th>GHSV (hr$^{-1}$)</th>
<th>Temperature (°C)</th>
<th>Conversion (%)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ce-Mo/TiO$_2$</td>
<td>Coprecipitation</td>
<td>19,000</td>
<td>200–400</td>
<td>90</td>
<td>[177]</td>
</tr>
<tr>
<td>MnAl/LDO</td>
<td>FNP</td>
<td>60,000</td>
<td>150–250</td>
<td>100</td>
<td>[178]</td>
</tr>
</tbody>
</table>
The powder catalyst in the laboratory stage is quite mature, meaning it can efficiently remove nitrogen oxides from simulated flue gas. However, how to apply these excellent catalysts to practice is a question worthy of consideration. Obviously, powder catalysts must be prepared as monolith catalysts to meet the needs of stationary industrial installations. Monolith catalysts have many dominant positions, such as strong mechanical stability, thermal conductivity, mass transfer capacity, small pressure drop and recycling, which are conducive to the catalytic process and practical application.

Yu Feng research group of Shihezi University has studied the monolith catalyst deeply. Tian et al. [185] prepared nanoporous microspheres Mn–Ce–Fe–Ti mixed oxide catalysts for NH₃-SCR by spray drying (Figure 11a). The samples were processed by focused ion beam (FIB) and observed by scanning electron microscopy (Figure 11b,c). After application in flue gas treatment, it was found that the monolith catalyst still maintained good catalytic activity. Wang et al. [159] first designed and prepared spherical MnOₓ-CeO₂-Al₂O₃ powder catalysts by spray drying, then applied it to monolith honeycomb catalysts (MHC). Compared with the same metal oxides catalysts prepared by coprecipitation (CP-MHC), the spray-drying (SD-MHC) method exhibited excellent SCR denitrification performance at 50–150 °C (Figure 11d,e).

<table>
<thead>
<tr>
<th>Catalyst Composition</th>
<th>Method</th>
<th>Temperature Range</th>
<th>Activity</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>MnOₓ-CeO₂-Al₂O₃</td>
<td>self-propagating synthesis</td>
<td>15,384</td>
<td>50–400</td>
<td>100</td>
</tr>
<tr>
<td>Mn–Ce-Ti</td>
<td>Hydrothermal</td>
<td>64,000</td>
<td>150–400</td>
<td>98</td>
</tr>
<tr>
<td>MnOₓ-CeO₂-TiO₂</td>
<td>Sol-ge</td>
<td>10,000</td>
<td>100–300</td>
<td>90</td>
</tr>
<tr>
<td>Co/Ni-CeO₂</td>
<td>Coprecipitation</td>
<td>48,000</td>
<td>75–200</td>
<td>93</td>
</tr>
<tr>
<td>Fe-Mn–Ce/γ-Al₂O₃</td>
<td>Sol-ge</td>
<td>10,000</td>
<td>100–450</td>
<td>95</td>
</tr>
<tr>
<td>MnOₓ/CeO₂-ZrO₂-Al₂O₃</td>
<td>Impregnation</td>
<td>10,000</td>
<td>50–300</td>
<td>90</td>
</tr>
</tbody>
</table>

4.2.2. CO-SCR

CO-SCR denitrification technology can effectively remove NO and CO from flue gas, thus performing two steps simultaneously [186]. In addition, CO-SCR as reducing agent can simultaneously remove NO and CO from tail gas and reduce the cost of purchase, transportation and storage of additional reducing agent. It is an attractive and promising denitrification technology.
Therefore, it is urgent to study CO-SCR technology [187–189]. Besides, precious metals are expensive, have limited reserves and have poor high temperature performance. They sinter at about 800 °C. At the same time, they have poor sulfur and phosphorus resistance, so it is particularly important to develop non-precious metal catalysts. Current research focuses on rare earth elements and cheap transition metals, rare earth elements, especially Ce, are widely studied in cheap transition metals, such as Cu, Fe, Mn, etc. [190–192].

In order to further explore the performance of CO-SCR denitrification, Shen et al. [187] found that the operating temperature window moved towards low temperature after Fe co-doping with Mn–Ce/TiO2. Yang et al. [193] studied the application of gas sensor in denitrification. Chen et al. [194] prepared Pd/LaFeO3 nanoparticle catalysts supported on LaFeO3 by two-step precipitation-deposition method. The visible light-induced valence exchange of Fe2+ and Fe3+ can promote the formation of oxygen vacancies. Lopes et al. [196] synthesized Cu-Ni catalyst by coprecipitation method. Bánsági et al. [197] found that isocyanate was the intermediate in the process of CO-SCR reaction. Wang et al. [198] studied the reaction of CO-SCR and the decomposition of supported metals by TG-FTIR. Liu et al. [190] prepared Cu0.1La0.1Ce0.8O catalyst, which had good water resistance and catalytic stability in CO-SCR process. Yamashita et al. [199] studied the CO-SCR reaction of nickel species supported on silica. Yao et al. [200] discussed the mechanism of CeO2 modified MnOx catalyst at low temperature (Figure 12a). Phase interfaces or synergies were formed on the surface (Figure 12b). Jin et al. [201] further studied the sulfur resistance and water resistance of Mn-Mo-WOx/TiO2-SiO2 catalysts. It was found that the catalytic activity of Mn-Mo-WOx/TiO2-SiO2 catalysts decreased significantly when H2O and SO2 existed simultaneously (Figure 12c,d), and the effects of different synthesis methods on NO conversion were compared in Table 5.

![Figure 12](image)

**Figure 12.** (a) The mechanism of selective catalytic CO removal from CeO2-MnOx-Al2O3 surface, (b) NO conversion of Ce-Mn/Al2O3 catalyst [200] (Reproduced from Ref. [200], Copyright 2016, Elsevier.), and (c,d) Mn-Mo-WOx/TiO2-SiO2 catalyst CO-SCR for sulfur and water resistance 201]. (Reproduced from Ref. [201], Copyright 2018, Elsevier.)

**Table 5.** Effects of different synthetic methods on de-NOx activity of CO-SCR.
### Catalyst Synthetic Method Reaction Conditions Temperature (°C) NO Conversion (%) Ref.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Synthetic Method</th>
<th>Reaction Conditions</th>
<th>Temperature (°C)</th>
<th>NO Conversion (%)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu0.1La0.1Ce0.8O</td>
<td>Grind</td>
<td>500 ppm NO, 1000 ppm CO, GHSV 26,000 h⁻¹, Ar</td>
<td>250</td>
<td>99.2</td>
<td>[190]</td>
</tr>
<tr>
<td>Mn–Ce/TiO₂</td>
<td>wetness impregnation</td>
<td>600 ppm NO, 1200 ppm CO, GHSV 40,000 h⁻¹, N₂, 200</td>
<td>94.9</td>
<td>[187]</td>
<td></td>
</tr>
<tr>
<td>Pd/CeZrO₂</td>
<td>Impregnation</td>
<td>CO, GHSV 24,000 h⁻¹, He</td>
<td>300</td>
<td>100</td>
<td>[194]</td>
</tr>
<tr>
<td>Pd/LaFeO₃</td>
<td>Two-step precipitation</td>
<td>5% NO, 10% CO, Water 10%, GHSV 24,000 mL h⁻¹, He</td>
<td>120</td>
<td>98</td>
<td>[195]</td>
</tr>
<tr>
<td>Cu-Ni/LDH</td>
<td>Precipitation</td>
<td>1.5 vol.% CO, 0.2 vol.% NO, O₂, 12,000 h⁻¹, He</td>
<td>400</td>
<td>100</td>
<td>[196]</td>
</tr>
<tr>
<td>Fe₀.₈Co₀.₂/ASC</td>
<td>Precipitation</td>
<td>500 ppm NO, 1000 ppm CO, GHSV 40,000 h⁻¹, N₂</td>
<td>200</td>
<td>96</td>
<td>[198]</td>
</tr>
</tbody>
</table>

#### 4.2.3. HC-SCR

In recent years, selective catalytic reduction of hydrocarbons (HC-SCR) has become a hot research topic worldwide. It has potential advantages in large-scale flue gas emission control, diesel engine and lean combustion engine exhaust gas treatment. At present, many studies have focused on Ag/Al₂O₃ catalytic system. Compared with the transition metal catalysts represented by Cu, Ag/Al₂O₃ catalysts have higher NO conversion and N₂ selectivity, outstanding thermal stability, water sulfur resistance, which makes it to be one of the most promising catalysts for HC-SCR.

H₂ can significantly promote the HC-SCR by hydrocarbons. Xu et al. [202] studied the sulfur resistance of Ag/Al₂O₃ catalyst assisted by H₂. It was found that Ag/Al₂O₃ catalyst with higher Ag loading had better denitrification and sulfur resistance, especially 4% Ag/Al₂O₃ catalyst, which was attributed to the rapid migration of sulfate on its surface to form more active sites. Thomas et al. [203] found that in the presence of H₂, the stability of nitrate was greatly reduced, and H₂ greatly reduced the temperature of formation and decomposition of NOₓ. Xu et al. [204] found that Ag/Al₂O₃ catalyst had excellent water resistance in the whole temperature range. After introducing water vapor, especially at low temperature, the conversion of NOₓ was enhanced. Chaieb et al. [205] studied the reaction mechanism of H₂ assisting C₃H₆ to remove NOₓ on Ag-loaded Al₂O₃ surface (Figure 13a). When the Ag-loaded density reached 0.7Ag/nm²Al₂O₃, the contact process between NO and C₃H₆ was the decisive step of C₃H₆-SCR process (Figure 13b). Xi et al. [206] prepared Fe-Ag/Al₂O₃ catalyst loaded on cordierite by sol–gel method and impregnation method. After Fe modification, the catalyst surface became loose and porous, forming Fe₃O₄-based needle-like and flaky crystals. It was found that Fe could effectively improve the performance of the catalyst against SO₂ and H₂O (Figure 13c,d), and increased the catalyst Lewis acid sites.
Figure 13. (a) The mechanism of C$_3$H$_6$ selective catalytic NO removal on Ag/Al$_2$O$_3$ surface, (b) the effect of Ag loading on NO conversion [205], and (c,d) the effect of Fe doping on the sulfur resistance and water resistance of Ag/Al$_2$O$_3$ catalyst [206]. Reproduced from Ref. [206], Copyright 2017, Elsevier.

4.2.4. H$_2$-SCR

H$_2$ is the most clean and resource-rich reductant for selective catalytic removal of NO$_x$. Compared with traditional NH$_3$ and urea as reductant, hydrogen as reductant has the advantages of low reaction temperature, clean and no secondary pollution, and low temperature activity of H$_2$-SCR catalyst.

Platinum and palladium, as unique active components, have excellent denitrification performance in the H$_2$-SCR system. Wang et al. [207] prepared Pt/HZSM-5 catalyst by impregnation method. FTIR analysis showed that Pt on the outer surface of molecular sieve acted as an activation agent for H$_2$ in H$_2$-SCR. Xue et al. [208] directly deposited Pt with different content on MIL-96 (Al) surface by hydrothermal method. Pt was slightly loaded on MIL-96 (Al) surface and H$_2$-SCR catalytic activity was excellent at 80 °C. Zhang et al. [209] modified H$_2$-SCR on Pt/HZSM-5 catalyst by tungsten doping. The results showed that tungsten was beneficial to the metal state of platinum in the catalyst and inhibited the formation of nitrate. At the same time, tungsten accelerated the dissociation of nitric oxide on the platinum surface. Väliheikki et al. [210] found that tungsten doping could improve the catalytic activity through the acidity of Ce$_2$Zr$_{1-x}$O$_2$ surface on H$_2$-SCR catalyst. Yang et al. [211] studied the catalytic performance of platinum supported on H-ferrous acid. In situ infrared spectroscopy showed that NO$_x$ and nitrate were not important intermediates of H$_2$-SCR on the catalyst, while NO$_2$ on Pt was an effective material for NO removal. Huai et al. [212] studied the decomposition process of NO on Pd (111) surface and the formation pathways of N$_2$, NH$_3$, N$_2$O, and H$_2$O (Figure 14a,b). The specific temperatures of them could be obtained by relative selectivity analysis at different temperatures (Figure 14c,d).
Figure 14. (a) Reaction pathways of NO + H2 and products on Pd (111), (b) potential energy diagram and geometrical structure of H atom-assisted direct dissociation of NO on Pd (111), and (c,d) relative selectivity of N2, NH3, NO, and H2O at different temperatures [212]. Reproduced from Ref. [212], Copyright 2015, Elsevier.

Removal of NOx by reduction is an extraordinary efficient method for removing NOx. However, SCR is limited by a series of problems, such as high cost, high temperature at 300 °C, NH3 escaping from secondary pollution environment [213], poor sulfur and water resistance of catalysts and susceptibility to poisoning and inactivation [214,215], which affect the life of the catalysts, makes the treatment and operation cost significantly higher [216,217]. Therefore, how to promote the resource-saving and environmentally friendly denitrification process is a difficult problem to be solved urgently in the world [218]. Studying the reaction mechanism of NO on catalyst surface is of great significance for solving the problem of catalyst poisoning and deactivation; improving catalytic denitrification activity; promoting dust, H2O, and SO2 resistance; enlarging operation temperature window; and promoting the low temperature direction of reaction [219–221].
5. Removal of NOx by Plasma

Plasma was discovered by German physicist and chemist Krux in 1879 and was first introduced into physics by American scientists Langmuir and Tonks. The plasma is the fourth form of matter, which contains a large number of electrons, ions, molecules, neutral atoms, excited state atoms, photons, and free radicals, among which positive and negative ions have the same charge [222]. Strictly speaking, plasma is a gas mass with high potential kinetic energy. The total charged quantity of plasma is still neutral. The outer electrons are shot out by the high kinetic energy of electric field or magnetic field. As a result, electrons are no longer bound to the nucleus, but become free electrons with high potential and high kinetic energy [223,224]. The plasma is a good conductive fluid, and there is no net Coulomb force or net magnetic force between charged particles. The charged particles are positive and negative in microscopy and electrically neutral in macro, and have certain thermal effects. According to the different thermodynamic equilibrium states, plasma can be divided into thermodynamic equilibrium plasma and non-thermodynamic equilibrium plasma [225,226]. Thermodynamically balanced plasma, also known as high temperature plasma, has the same temperature of molecule, electron and ion [227]. Non-thermodynamic equilibrium plasma is also called cryogenic plasma, in which the electron temperature is 10,000–100,000 K, the temperature of ions and neutral particles is much lower than that of electrons, the temperature is only 300–500 K, and the temperature of the whole system is lower [228–230]. Low temperature plasma has been widely used in many fields due to its unique characteristics [231]. According to the treatment of NOx by plasma, it can be divided into three parts: direct decomposition of NOx by plasma, plasma-modified catalyst and plasma assisted catalyst.

5.1. Direct Decomposition of NOx by Plasma

The plasma can not only directly decompose organic compounds in sewage, but treat nitrogen oxides directly [232]. The study of plasma direct decomposition of NOx dated back to 1997. T.Oda et al. [233] used dielectric barrier discharge to decompose NOx in automobile exhaust directly. They found that NOx was more easily oxidized in the presence of oxygen. Baeva et al. [234,235] also found the same phenomenon when he decomposed NOx by pulsed microwave discharge plasma at atmospheric pressure. Hueso et al. [236,237] used microwave-induced plasma to study the decomposition of NO in Ar and N2. In the presence of oxygen, if enough reductive gases were added, the decomposition of NO was still dominant. Schütz et al. [238] introduced that dielectric barrier discharge can be realized in air atmosphere, discharge uniformly, produce high concentration of isomers, and can be effectively utilized. There are many factors affecting dielectric barrier discharge, such as material of dielectric barrier, discharge voltage, discharge frequency, discharge distance, etc. [239]. Wu et al. [240] proposed a new method of corona discharge coupled with wet absorption, which can achieve simultaneous desulfurization and denitrification. Chen et al. [241] studied the reaction pathway and mechanism of NO removal by nonthermal plasma NTP for the first time by FT-IR (Figure 15a). It was found by FT-IR analysis and recording that O· played the role of NO· + O· → NO2 oxidation (Figure 15b). The effect of plasma on NO removal in Ar, O2, N2, and other atmospheres was compared. The sulfur and water resistance were also studied, NO3− and SO42− were formed as the byproducts, which greatly attenuate the catalytic activity (Figure 15c,d).
5.2. Plasma-Modified Catalyst

NO\textsubscript{x} removal by plasma-modified catalysts is a new green and environmental technology. The modification of catalysts by plasma is mainly focused on the surface modification of catalysts. The dispersion of active species can be improved by plasma modification, and the stability and low temperature activity of catalysts can be improved [242]. Hong et al. [243] studied the denitrification performance of pulsed corona discharge plasma. The results showed that there was an obvious interaction between the process of desulfurization and denitrification. Zhang et al. [244] treated NiO-TiO\textsubscript{2}-Al\textsubscript{2}O\textsubscript{3} catalyst with plasma. It was found that the specific surface area of the catalyst was increased, the nickel particles dispersed more evenly and the ability of the catalyst to adsorb NO\textsubscript{x} was enhanced. Huang et al. [245] prepared V\textsubscript{2}O\textsubscript{5}/ACF catalyst. After oxygen plasma modification, the surface oxygen-containing functional groups increased, the pore size distribution widened and the catalytic activity increased. Liu et al. [246] prepared a Mn-O-Ce catalyst by nonthermal plasma, it decomposes nitrate and organic matter slowly and forms defective Mn-O-Ce phase in the catalyst during plasma treatment. More details are shown in Table 6.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Conversion (%)</th>
<th>Temperature (°C)</th>
<th>Ref.</th>
</tr>
</thead>
</table>
| NiO-TiO\textsubscript{2}-Al\textsubscript{2}O\textsubscript{3} | Without plasma < 12  
Plasma-modified > 70 | 180–240 | [244] |
| V\textsubscript{2}O\textsubscript{5}/ACF | Without plasma < 35  
Plasma-modified > 55 | 50–150 | [245] |

The plasma contains high energy, which can provide enough activation energy for many difficult reactions. An et al. [247] studied the oxidation characteristics of NO in oxygen atmosphere by surface dielectric barrier discharge plasma treatment of mixed flue gas with active material injection method. Cui et al. [248] prepared a MnCe/Ti catalyst in a dielectric barrier discharge reactor by combining nonthermal plasma discharge with catalyst. The highest NO conversion rate was 86.9%. Zhao et al. [249] developed a new process of denitrification by water-cooled dielectric barrier
discharge. Peng et al. [250] used nonthermal plasma and bamboo charcoal combined with adsorption technology to catalyze the removal of NO. SEM showed that the surface of the catalyst after plasma treatment would produce defects to promote the growth of pore structure and increase the active sites. Park et al. [251] removed both NO and SO2 from flue gas through a wet NaClO2 scrubber combined with a plasma electrostatic precipitator. Wang et al. [252] prepared Mn–Ce/ZSM5 multi-walled carbon nanotubes catalyst by impregnation (Figure 16a). It was found that more oxygen holes could be generated after plasma treatment, which could significantly improve NO conversion (Figure 14b). Among them, MnCe/ZSM5 has the highest catalytic activity and its reaction accorded with Eley–Rideal mechanism (Figure 16c). The sulfur and water resistance of MnCe/ZSM5 were also slightly improved (Figure 16d).

**Figure 16.** (a) Mechanism of NOx removal by plasma-modified catalyst Mn–Ce/ZSM5, (b) effect of different Mn–Ce ratios and plasma treatment on NO conversion, (c) denitrification process of catalyst Mn1Ce1/ZSM5 conforms to Eley–Rideal mechanism, and (d) study on sulfur and water resistance of plasma-modified catalyst [252]. Reproduced from Ref. [252], Copyright 2018, Elsevier.

5.3. Plasma Concerted Catalysis

Plasma concerted catalysis is the combination of plasma and catalyst, which makes them act synergistically on the reaction system and optimize the reaction process. According to the combination of catalysts and plasma, plasma-assisted catalysis can be divided into plasma-enhanced catalysis and plasma-driven catalysis. (1) Plasma-enhanced catalysis: catalysts work at the back of the plasma and the plasma and catalyst work in the optimum temperature range respectively. Under this combination mode, the role of plasma is to change the gas composition reaching the catalyst bed. Reactive gases are treated by plasma to produce more highly active intermediates, which react on catalysts to produce final products [253]. (2) Plasma-driven catalysis: plasma and catalyst are placed in the same reactor, and a large number of short-lived active species produced by plasma change the shape of gaseous reactants on the catalyst. The discharge of plasma on the catalyst surface may also change the properties of the catalyst, such as the generation of electron holes and electrons, and the surface of the catalyst. The change of work function, the activation of lattice oxygen of catalyst, the formation of new active sites on catalyst surface, etc. [254]. Tang et al. [255] studied the effect of nonthermal plasma co-catalytic treatment on the low-temperature oxidation of NO by Mn-CoOx.
catalyst (Figure 17a,b). It was found that the activity of the catalyst treated by nonthermal plasma increased significantly (Figure 17c), and the specific surface area and pore volume of the catalyst increased significantly after nonthermal plasma treatment, as well as the advantages of changing the relative surface concentration and oxidation state of the species on the catalyst surface. Two main weightlessness peaks were studied (Figure 17d).

**Figure 17.** (a) Schematic diagram of plasma-enhanced catalysis and plasma-driven catalysis, (b) mechanism of low-temperature denitrification of nonthermal plasma-enhanced Mn-CoOx catalyst, (c) effect of plasma voltage and treatment time on NO conversion, and (d) study on thermal stability of thermogravimetric curves [255]. Reproduced from Ref. [255], Copyright 2015, Elsevier.

To further demonstrate the concerted effect of plasma, the following datas are listed to enhance persuasion. Niu et al. [256] found that the denitrification rate was greatly increased when the plasma acted with Co-ZSM-5 zeolite catalyst. Bröer et al. [257] found that the activity of SCR at low temperature increased significantly under the action of plasma with V₂O₅-WO₃/TiO₂ catalyst. Fan [258] studied the concerted effect of plasma and H₂ mordenite in C₂H₂-SCR. Li et al. [259] explored the enhanced effect of Mn-Co-CeOₓ catalyst with plasma treatment in NH₃-SCR. Cho et al. [260] discussed the concerted effect in plasma-enhanced catalysis. Using Ba & Cu (C₆H₁₂O₃) as catalyst, NO was oxidized to NO₂ in plasma, while hydrocarbon was oxidized to active intermediate species CO, which eventually reduced NOₓ to N₂. Pan et al. [261] found that the conversion of NOₓ as well as the water and sulfur resistance of C₂H₂-SCR plasma increased significantly when dielectric barrier plasma acted simultaneously. Wang et al. [262,263] found that the denitrification rate was very low when the plasma acted alone. When the plasma cooperated with the Mn-Cu/ZSM5 zeolite catalyst, the conversion of NO was greatly increased. The above data are summarized in Table 7.
Table 7. Effect of plasma addition on denitrification activity of different catalysts.

<table>
<thead>
<tr>
<th>Catalyst</th>
<th>Conversion (%)</th>
<th>Temperature (°C)</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co-ZSM-5</td>
<td>Plasma only 12</td>
<td>150–450</td>
<td>[256]</td>
</tr>
<tr>
<td></td>
<td>Catalyst only 85</td>
<td>300</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Plasma assist catalyst 95</td>
<td>300</td>
<td></td>
</tr>
<tr>
<td>V₂O₅-WO₃/TiO₂</td>
<td>Plasma only 20</td>
<td>150–260</td>
<td>[257]</td>
</tr>
<tr>
<td></td>
<td>Catalyst only 35</td>
<td>180–260</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Plasma assist catalyst 80</td>
<td>160–260</td>
<td></td>
</tr>
<tr>
<td>H-mordenite</td>
<td>Plasma only 3.8</td>
<td>200</td>
<td>[258]</td>
</tr>
<tr>
<td></td>
<td>Catalyst only 54</td>
<td>200</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Plasma assist catalyst 91.4</td>
<td>200</td>
<td></td>
</tr>
<tr>
<td>Mn-Co-CeO₂</td>
<td>Plasma only 73</td>
<td>150</td>
<td>[259]</td>
</tr>
<tr>
<td></td>
<td>Catalyst only 70</td>
<td>150</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Plasma assist catalyst 93</td>
<td>100</td>
<td></td>
</tr>
<tr>
<td>Ba &amp; Cu (C₂H₂O₃)₂</td>
<td>Plasma only 85</td>
<td>25–400</td>
<td>[260]</td>
</tr>
<tr>
<td></td>
<td>Catalyst only 82</td>
<td>25–400</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Plasma assist catalyst 95</td>
<td>200</td>
<td></td>
</tr>
<tr>
<td>Mn–CoOₓ</td>
<td>Plasma only 3.8</td>
<td>200</td>
<td>[255]</td>
</tr>
<tr>
<td></td>
<td>Catalyst only 54</td>
<td>200</td>
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</tr>
<tr>
<td></td>
<td>Plasma assist catalyst 91.4</td>
<td>200</td>
<td></td>
</tr>
<tr>
<td>Mn-Cu/ZSM5</td>
<td>Plasma only 50</td>
<td>25–450</td>
<td>[262]</td>
</tr>
<tr>
<td></td>
<td>Catalyst only 60</td>
<td>25–450</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Plasma assist catalyst 90</td>
<td>25</td>
<td></td>
</tr>
</tbody>
</table>

In recent years, the form of plasma has been innovated rapidly [264,265]. Dielectric barrier discharge, radio frequency discharge, and microwave discharge are all new nonthermal equilibrium plasma discharge forms [266,267]. The effect of plasma denitrification is remarkable, which can improve the NO conversion rapidly on short notice. However, compared with the traditional denitrification technology, the plasma process has made some progress, but there are still some problems: high energy consumption, short power supply life and performance to be improved; moreover, the expensive price of plasma equipment, high cost of system operation and maintenance, and complex equipment structure make the technology not widely used and restrict its development.

6. Conclusions and Prospect

Along with the huge global emission of NOₓ, which continues to deteriorate planet earth environment and human health in the Anthropocene, it becomes progressively significant to collect and summarize the efficient approaches of governance and treatment of NOₓ. Optimizing the above four distinct denitrification methods and controlling the emission of NOₓ are the main challenges for future atmospheric environmental treatment. Finally, in order to solve the problem of air pollution that plagues all countries in the world, the feasibility of four main technologies in the field of flue gas denitrification and industrial application of flue gas tail purify is evaluated.

Adsorption treatment of NOₓ has the features of environmental friendliness, energy-saving, and high efficiency. The process is simple to operate, controllable, for low concentration poisonous gases, efficient capture can be achieved. However, due to their limited adsorption capacity, frequent regeneration of adsorbents, and huge size and the large-scale investment required, the application of adsorbents is limited. Oxidation process is a relatively mature method in the field of wet denitrification technology because of its simple route, easy operation and outstanding denitrification effect. Some of them have been industrialized. However, the oxidation tower has the problems of expensive raw materials and equipment corrosion, which limits its development due to its cost and safety. Reduction is the most efficient and mature denitrification technology with the most extensive application as well as the most exceptional means to control NOₓ pollution. But at the same time, it
has some shortcomings, high investment, high cost of catalyst regeneration, NH₃ escaping from secondary pollution, and poor sulfur and water resistance of catalyst and so on. The removal of NOₓ by plasma technology is remarkable, which can rapidly improve the denitrification performance in a short time, and promote the stability and low temperature activity of the catalyst. However, the required equipment covers a large area, consumes high energy, and costs high investment, operation and maintenance, which is limited in practical application.

Future, the general requirement of denitrification technology is low cost, high efficiency and green. The overall development trend of technology is to realize the coordinated removal of multiple pollutants by coupling multiple technologies. Different regions and industries are suitable for different denitrification means. In order to decrease the emission level of NOₓ, cut down the expense of treatment and recovery, and improve the economic performance, different regions and industries should choose appropriate denitrification technology according from the resource situation to product usage. Therefore, it is of great practical significance to develop denitrification methods with high efficiency, low energy consumption, low secondary pollution, and low investment.

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