MINIREVIEW

A review on oxidation of elemental mercury from coal-fired flue gas with selective catalytic reduction catalysts

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Catalytic technologies present a more environmentally and financially sound option in the removal of elemental mercury (Hg^0) from coal-fired flue gas. However, developing novel and efficient catalysts for Hg^0 oxidation is still a challenge. This paper reviews the catalytic oxidation of Hg^0 over a new kind of catalysts which were developed from selective catalytic reduction (SCR) catalysts of NO_x. In this review, both noble metal catalysts and non-noble metal catalysts for Hg^0 oxidation were summarized. An overview of mercury emissions including transformation and speciation of mercury in coal-fired flue gas was also presented. The possible mechanisms and kinetics of mercury oxidation, space velocity and the effects of flue gas components on activity and stability of the catalysts were examined. We expect that this work will serve as a theoretical underpinning for the development of Hg^0 oxidation technology in flue gas.

1 Introduction

Mercury (Hg) has been known for a long time to be an environmental contaminant which is toxic to human beings and other organisms. It is well known that, in anthropogenic activities, coal combustion is a major source of Hg emissions.[1] Due to the harmful environmental impacts, Hg emission legislation becomes increasingly stringent. In 2013, the US Environmental Protection Agency (USEPA) updated the

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emission limits of Mercury and Air Toxics Standards (MATS), which requires that the total emission of mercury from new coal-fired units burning low rank virgin coal must be controlled below the level of 0.003 lb GWh^{-1}.[2,3] Furthermore, in January 2013, 140 nations adopted the first legally binding international treaty to set enforceable limits on emissions of Hg and exclude, phaseout, or restrict some products that contain Hg.[4]

To meet the stringent regulation, various technologies for controlling Hg emissions, including adsorption technology, corona discharge plasma technology, and electrocatalytic oxidation combined treatment technology, have been investigated.[5-7] Unfortunately, it is difficult to apply these technologies widely because of the unaffordable cost. Hence, developing a low cost

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Caiting Li

option for controlling Hg emissions from coal-fired power plants is indispensable. Due to the strict regulations for air pollution, most of the coal-fired power plants have been equipped with air pollution control devices (APCDs), such as fabric filters (FFs) and electrostatic precipitators (ESPs) for particulate control, wet flue gas desulfurization (WFGD) for SO_2 control and selective catalytic reduction (SCR) for NO_x emission control.[8] In flue gas, Hg primarily exists in three forms: elemental mercury (Hg^0), oxidized mercury (Hg^{2+}), and particle-associated mercury (Hg^p). It has been reported that the existing APCDs can achieve the co-benefits of Hg capture.[9,10] For example, Hg^p can be collected by ESPs and FFs together with fly ash.[11] Highly water-soluble Hg^{2+} might be effectively captured by WFGD.[12–14] However, it is difficult to remove Hg^0 directly by existing APCDs because it's highly volatile and nearly insoluble in water.[15–18] Therefore, a combination of Hg^0 oxidation and WFGD is considered as an effective option for Hg emission control.[19–21]

SCR catalysts for NO_x removal were proved to be able to oxidize Hg^0 to Hg^{2+} and lots of full-scale tests were carried out to evaluate the performance of these SCR catalysts in Hg^0 oxidation.[22–24] It is found that a combination of ESP, SCR and FGD is effective in removing appreciable levels of Hg^0.[19] Overall mercury removal efficiencies of APCDs, on average, were about 61% and 47% with and without an SCR system, respectively.[22] Blythe[25] compared the cost of catalytic oxidation technology and activated carbon injection (ACI). It was proved that the co-benefit effect of the SCR system for oxidizing Hg^0 makes the cost of Hg^0 removal lower than that of ACI. Hence, combining the SCR system with WFGD is thought to be one of the most economic approaches for controlling Hg emissions from coal-fired power plants. Some studies on researching selective catalytic reduction of NO_x and Hg^0 removal have been done by our group.[17,26–29]

Understanding the transformation and speciation of Hg throughout the coal-fired process is crucial to the design of effective technologies for Hg^0 removal. The oxidation of Hg^0 is helpful to obtain greater mercury capture efficiency with the APCDs. Accordingly, this paper introduces the research progress on Hg^0 oxidation over a new kind of catalysts which were developed from selective catalytic reduction (SCR) catalysts of NO_x. The transformation and speciation of Hg in coal-fired flue gas will be considered first, followed by a review on the mechanistic pathways and kinetics of mercury oxidation. At last, the influence of flue gas components, space velocity and temperature is summarized and reviewed.

2 Mercury emissions in flue gas

2.1. Mercury emissions

After the 19th century, a large amount of Hg is emitted into the environment due to anthropogenic activities, leading to considerably increasing Hg levels in the atmosphere.[30] According to the United Nations Environment Programme (UNEP), the global Hg emissions to air from anthropogenic sources were estimated to be 1960 tonnes in 2010.[31] Coal combustion, waste incineration, base metal smelting, large-scale and artisanal gold production and cement production are the most important anthropogenic sources of Hg emissions.[32,33] As listed in Table 1,[34] coal combustion is the biggest contributor to Hg emissions. As a big coal consumer, Hg emissions from coal-fired power plant can't be ignored and have been studied by many researchers. Streets *et al.*[35] estimated that China's emissions in 1999 were 536 (±236) tons, and approximately 38% of the Hg comes from coal combustion. At the same time, Wu *et al.*[36] also estimated that total Hg emissions from all anthropogenic sources increased at an average annual rate of 2.9% during the period 1995–2003, reaching 696 (±307) tons in 2003. The USEPA estimated that approximately 75 tons of Hg is found during the process of coal transportation in the United States each year and about two thirds of the mercury is emitted to the air.[37] Using South Africa specific and toolkit based emission factors, coal-fired power plants were estimated to be the largest contributor of Hg emissions, *viz.* 27.1 to 38.9 tonnes per year in air.[38]

2.2 Transformation and speciation of mercury in flue gas

In order to understand the transport and fate of Hg in air pollution control systems, it is necessary to investigate Hg transformations and speciation in coal-fired flue gas. Hg was found to be the most volatile element in coal. It can volatilize at temperatures as low as *c.a.*150 °C. The volatility of Hg increased with increasing temperature above 400 °C. Fikleman *et al.*[39] showed that the volatilization rate of Hg in the Argonne Premium Coal Samples is about 40–75% at 550 °C. Rizeq *et al.*[40] suggested that Hg could be volatilized completely when temperature was higher than 800 °C. Therefore, while entering the furnace, most of Hg is rapidly volatilized. It moves through the convective section and economizer of the boiler island before exchanging heat in the air pre-heater. With the temperature of flue gas decreasing, gaseous Hg is predicted to react with the component of flue gas. Eventually, the principal form of Hg in coal combustion flue gas is assumed to be Hg^0.[41] Fig. 1 presents the migration mechanism of mercury in the coal combustion process and
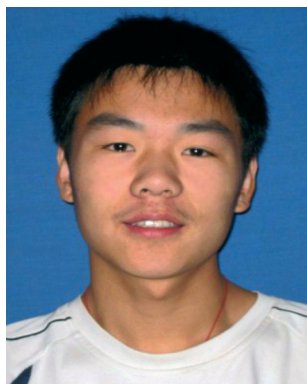
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3460 | *Catal. Sci. Technol.*, 2015, **5**, 3459–3472

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Table 1 Contribution of sources of anthropogenic mercury emissions expressed as a percentage (%) of total emissions (ref. 34)

Emission type		USA	China	Canada	Europe	Africa	Global
Coal combustion	Power plants	32.6	12.7	3.6	26.1	51.5	—
	Industrial	13.1	19.3	—	—	—	—
	Residential	—	3.7	—	26.1	0.1	—
	Total	45.7	35.7	3.6	52.2	51.6	65.0
Waste incineration[a]		33.8	1.1	9.4	2.8	—	3.0
Base metal smelting[b]		0.1	36.9	66.0	4.5	2	6.8
Gold production[c]		—	13.6	9.4	—	44.6	11.3
Mercury production		0.3	1.6	—	—	—	1.1
Chlor-alkali plants		4.5	0.04	1.4	12.1	0.1	3.0
Cement production		3.1	4.2	3.0	8.4	1.3	6.4
Iron & steel industry		—	—	0.8	3.4	0.1	1.4
Other		12.5	6.9	28.0	16.6	0.4	2.0

[a] Includes municipal, medical, sewage sludge and hazardous waste incineration. [b] Includes copper, lead and zinc smelting. [c] Includes artisanal and large-scale gold mining.

flue gas.[41] At furnace exit temperatures (1700 K), all mercury is expected to remain in the favoured elemental form of thermodynamics in the gas.[42] However, Hg^0 vapor undergoes several chemical and physical process changes in the post combustion section, where the gas temperature decreases. It reacts with other flue gas constituents to convert to gaseous Hg^{2+} and Hg^p as the temperature of flue gases falls down below 600 °C. Hence, the forms of Hg in coal-fired flue gas are Hg^0, Hg^{2+}, and Hg^p.[43,44]

Researchers have proposed different Hg reaction mechanisms to describe Hg transformations in coal-fired flue gas.[45–47] To date, it has been widely accepted that both heterogeneous and homogeneous reactions play important roles in mercury–flue gas chemistry. The fate of Hg species in coal flue gas is determined by the results of heterogeneous and homogeneous reactions occurring in utility systems. Meanwhile, the formation of various Hg species is affected by many parameters, including the component of flue gas, combustion environment and plant operating conditions. Therefore, some researchers focused on understanding the mechanisms of Hg oxidation by injection of Hg^0 into gas fuel flame or simulated flue gas. The experimental data obtained by

Bool *et al.*[48] indicated that most of mercury vaporized and either left the reactor as a vapor or was captured by residual carbon. The equilibrium predicted that HgO might form and condense on the ash. NO_2, HCl, and SO_2 exhibited a promotional effect on Hg^0 oxidation, while NO inhibited Hg^0 oxidation.[49] Nevertheless, the extent of homogeneous Hg^0 oxidation is highly dependent upon the coal rank, the content of Cl in the coal, and the conditions of the utility boiler (*e.g.*, air-to-fuel ratio and temperature).[50] Thermodynamic calculation has predicted that Hg will be in the form of Hg^0 and $HgCl_2$ at typical temperatures in flue dusts (80–250 °C). $HgCl_2$ is a stable mercury species followed by $HgO_{(g)}$ in a chlorine-laden flue gas at temperatures lower than 400 °C. Meanwhile, a literature survey revealed that Hg^0 oxidation occurs at temperatures below 700 °C and that mercury will be completely oxidized at (or below) 450 °C.[42] However, $Hg^0_{(g)}$ is the only thermodynamically stable species above 750 °C.[51] Consequently, Hg is mainly distributed in gaseous (Hg^0 and Hg^{2+}) form.[52] Generally, more than half of the gas phase Hg exists as Hg^{2+} which is likely to be $HgCl_2$ (50–80%), and the remaining is Hg^0 (20–50%).[53–55]

3. The catalytic oxidation of Hg^0 on SCR catalysts

The SCR technology for control of NO_x emissions from flue gas is the best developed and world-wide used technology since the 1980s.[56] The main overall reactions can be expressed as:

$$4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O \tag{1}$$

$$6NO_2 + 8NH_3 \rightarrow 7N_2 + 12H_2O \tag{2}$$
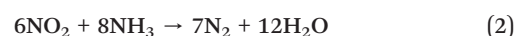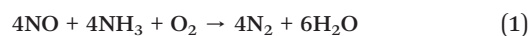
In addition to NO_x control, SCR catalysts exhibit the co-benefit of promoting Hg oxidation in coal-fired power plants.[19] Two types of catalysts have been developed, noble metal-based catalysts and non-noble metal-based catalysts. As summarized in Table 2, these two types of catalysts have
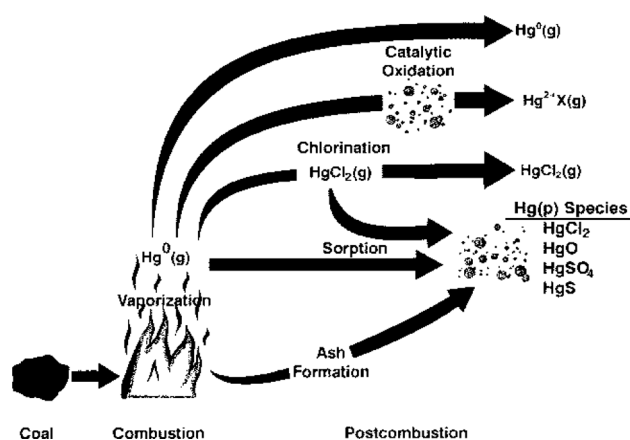

Fig. 1 Migration mechanism of mercury in coal combustion process and flue gas.[41]

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Catal. Sci. Technol., 2015, **5**, 3459–3472 | **3461**

Table 2 Elemental mercury oxidation on SCR-DeNO$_x$ catalysts

Catalyst type	Potential catalysts	Gas composition							T °C	Space velocity h^{-1}	Hg^0 oxidation %	Reference
		O$_2$ vol. %	H$_2$O vol. %	HCl ppm	NO ppm	NH$_3$ ppm	SO$_2$ ppm	Hg^0 µg N m^{-3}				
Noble metal-based catalysts	Pd/Al$_2$O$_3$	—	—	10	—	—	—	70	200–350		>90	57
	Au/TiO$_2$	4	10	50	100	—	1000	20–30	150	1200[a]	40–60	58
	Pd/Al$_2$O$_3$	0–5.25	—	0–100	500	—	0–1000	6–18	138–160	8–10[b]	1.6 × 10^{-10}[c]	61
Non-noble metal-based catalysts	V$_2$O$_5$/TiO$_2$	1.6	8	10	160	52.8	160	1.2[d]	250–400	170·	>90	74
	MnO$_x$/TiO$_2$	2	—	—	400	—	—	15–66[d]	175–200	5000	>90	12
	MnO$_x$-CeO$_2$/TiO$_2$	4	8	10	300	300	400	75	100–400	6 × 10^5	0–90	75
	Mo–Mn/α-Al$_2$O$_3$	7.1	6.8	0–20	400	—	500	—	100–250	4.4 × 10^4	70–100	11
	CeO$_2$-WO$_3$/TiO$_2$	8	8	10	—	—	500	80–100	100–500	1.0 × 10^5	>80	82
	CeO$_2$-TiO$_2$	4	8	10	300	—	400	50	120–400	6 × 10^5	>90	84
	Commercial SCR catalysts	6	—	50	400	400	—	36–39	350	4000	3–91	64
		—	15	0.3–3	400	300	70	160	260–320	170[a]	50–90	91
		3	—	500	250	275	2000	120	300–350	1800	<80	94

[a] Gas space velocity (L h^{-1}). [b] Gas space velocity (L min^{-1}); [c] Reaction rate in the presence of HCl and O$_2$ in (mol Hg^{2+}) × (g catalyst)$^{-1}$ × s^{-1}. [d] PPb.

been primarily studied for heterogeneous catalytic oxidation of Hg^0.

3.1 Noble metal-based catalysts

Noble metals, such as Au, Pd, Pt, and Rh, which are used as potential Hg^0 oxidation catalysts, have been used to test their Hg adsorption abilities because of their regeneration potential and good stability at high temperatures. In order to maximize specific surface areas of the catalysts, the noble metals used for the Hg^0 oxidation are supported by various porous materials, including alumina, silica, zirconia, titania, carbons, and zeolite. For example, a mass loading of 8% Pd supported by alumina can remove over 90% of mercury for operating temperatures up to 270 °C.[57] Au/TiO$_2$ was also effective, yielding Hg oxidation ranges of 40–60%.[58]

In particular, Pd has been considered as the most attractive option for controlling Hg^0 emissions.[59,60] In the study by Presto,[61] the Pd catalyst exhibited no apparent catalyst deactivation with HCl concentration changed. When the HCl concentration increases from 50 to 100 ppm, little impact on the Hg^0 oxidation rate was observed. In the absence of HCl, however, it was observed that Hg^0 oxidation still continues on the catalysts, while with a declining reaction rate. From this observation, it was suggested that the reactions between mercury and HCl are bound to the catalyst surface. This explains why Hg^0 oxidation continues in the absence of HCl, but with a declining reaction rate.

Au has been considered as a very promising candidate catalyst for Hg^0 oxidation because Au can adsorb and react with Hg^0 to form amalgam.[61,62] Lim et al.[62] suggested that adsorbed Hg^0 on the Au catalyst reacts with Cl$_2$ (or HCl) in accordance with the Langmuir–Hinshelwood mechanism. Activation energies for Hg oxidation were calculated assuming the possible reaction pathways: three-step Hg oxidation (Hg → HgCl → HgCl$_2$) with transition states TS$_1$ and TS$_2$

(Fig. 2). In this Hg^0 oxidation, the first Cl attachment step is exothermic, while the second Cl attachment step is endothermic. It is implied that Hg^0 oxidation prefers a pathway in which HgCl and HgCl$_2$ are formed, rather than a pathway directly oxidizing Hg to HgCl$_2$. Another literature mentioned a similar Hg^0 oxidation trend.[63] Atomic Cl is the key species for Hg^0 oxidation on the surfaces of gold. That is, Hg^0 first reacts with one atomic Cl to form HgCl, which, in turn, is oxidized to HgCl$_2$ by the other Cl.

3.2 Non-noble metal-based catalysts

Noble metal catalysts used as catalysts for SCR of NO in flue gas are still an area of active study. However, they are too expensive to apply in industry. Consequently, noble metal catalysts were soon replaced with non-noble metal catalysts for SCR of NO. Non-noble metal catalysts, especially some transition metal catalysts, have been observed to be beneficial to oxidize Hg^0 to Hg^{2+} when sufficient HCl exists in the
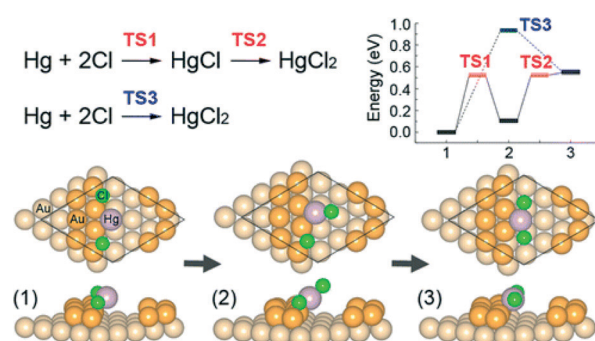

Fig. 2 Reaction pathways of Hg oxidation on perfect Au(111)–p(4 × 4) surfaces.[62] Reprinted (adapted) with permission from (D. H. Lim and J. Wilcox, Environ Sci Technol, 2013, 47, 8515–8522.). Copyright (2013) American Chemical Society.

3462 | Catal. Sci. Technol., 2015, 5, 3459–3472

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flue gas. Therefore, various metal catalyst materials have been investigated for Hg^0 oxidation in recent years.[11,64–67]

3.2.1 Transition metal oxide catalysts. To date, lots of research involving transition metal oxide catalysts, such as V_2O_5, MnO_2, Co_3O_4, CuO and TiO_2, have been extensively conducted to develop effective Hg^0 oxidation technologies.[68–72] Compared with noble metal catalysts, the lower-cost transition metal catalysts exhibit high catalytic oxidation activity. Transition metal oxide catalysts for Hg^0 oxidation are usually supported by various materials, including alumina, silica, titania, carbons, zeolite, etc. Generally, alumina and titania are used as carriers for these transition metal oxide catalysts. It is well known that the role of the supporter is not only to stabilize and ensure a high metal dispersion degree, but also in certain cases to participate in the Hg^0 oxidation reaction.[73] Kamata et al.[74] investigated Hg^0 oxidation by HCl over the metal oxides (1 w.t.% MO_x where M = V, Cr, Mn, Fe, Ni, Cu, and Mo) supported on anatase type TiO_2. The metal oxides added to the catalyst were observed to be dispersed well on the TiO_2 surface. Meanwhile, the catalyst such as V_2O_5/TiO_2 showed high NO reduction activity and high Hg^0 oxidation activity.

Among several metal oxide catalysts,[67,75] the manganese-based catalysts were found to be one of the best candidates for Hg^0 oxidation owing to their excellent catalytic activity, easy manufacturing and low cost. It has been studied extensively as low-temperature SCR catalysts because they possessed various types of labile oxygen which played an important role in the catalytic reaction.[76,77] Meanwhile, MnO_x based SCR catalysts can also serve as catalysts for Hg^0 oxidation. Ji et al.[12] reported that MnO_x supported on titania was effective for both elemental mercury capture and low temperature SCR. The results indicated that MnO_x/TiO_2 catalyst could achieve 97% NO conversion and capture approximately 90% of the incoming Hg. However, for manganese-based catalysts, the influence of SO_2 poisoning is a major problem. In order to further improve the sulphur tolerance of catalysts at low temperature, several metal elements (CeO_2, W, Mo) were employed as dopant to modify the manganese-based catalysts. The CeO_2 doped catalyst displayed excellent sulfur tolerance performance at low temperature.[78] In particular, Mn–Ce mixed-oxide exhibited an excellent Hg^0 removal capacity. Li et al.[75] found that the combination of MnO_x and CeO_2 resulted in significant synergy for Hg^0 oxidation. The Mn–Ce/Ti catalyst was highly active for Hg^0 oxidation at low temperatures (150–250 °C) under both simulated flue gas and SCR flue gas (see Fig. 3). The Mo doping also resulted in high Hg^0 oxidation in gases with 5ppm HCl, even in the presence of SO_2.[11]

As is well known, cerium has received considerable attention due to its prominent ability to store/release oxygen as an oxygen reservoir via the redox shift between Ce^{4+} and Ce^{3+} under oxidizing and reducing conditions, respectively.[17,79] Besides, cerium oxide-based catalysts were reported to have good resistance to water vapor.[80] And the doping of CeO_2 greatly enhanced the SO_2 resistance of the catalyst.[81]
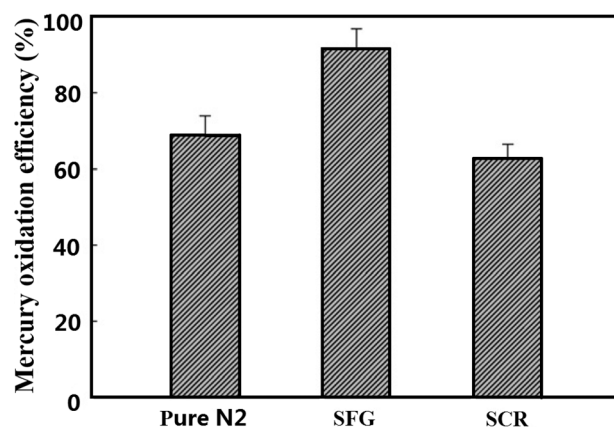

Fig. 3 Hg^0 oxidation under different atmospheres at 200 °C.[75]

Therefore, cerium oxide is considered as a very promising candidate for mercury oxidation. Wan et al.[82] studied the removal of Hg^0 over a CeO_2-WO_3/TiO_2 nano-composite in simulated coal-fired flue gas. About 95% of the Hg^0 could be removed by HCl in the presence of O_2. The Hg^0 removal efficiency was found to be slightly affected by H_2O addition, while SO_2 promoted the Hg^0 oxidation. Remarkably, the CeO_2-TiO_2 materials exhibited excellent single and simultaneous capture capacities.[81] Most likely cerium can occupy two oxidation states [CeO_2 (Ce^{4+}) $\leftrightarrow$ Ce_2O_3 (Ce^{3+})], allowing ceria from the CeO_2-TiO_2 support to accommodate more surface lattice oxygen species. Consequently, Hg^0 adsorbed on the ceria surface can react with the lattice oxygen to form HgO. Zhou et al.[83] believed that Hg^0 oxidation over CeO_2-TiO_2 catalysts could be explained by the Mars–Maessen mechanism, in which active surface sulfur reacts with gas-phase Hg^0. The possible mechanism is proposed in Fig. 4. However, the research by Li et al.[84] showed different results. It proposed that Hg^0 oxidation over CeO_2-TiO_2 catalysts followed the Langmuir–Hinshelwood mechanism whereby reactive species from adsorbed flue gas components react with adjacently adsorbed Hg^0.

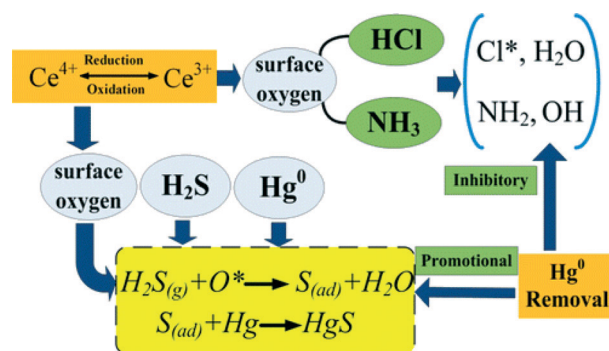**3.2.2 Commercial SCR catalysts.** In recent years, the SCR system has been extensively used in coal-fired plants to

Fig. 4 Mechanism of CeO_2-TiO_2 catalysts for elemental mercury removal.[83] Reprinted (adapted) with permission from (J. Zhou, W. Hou, P. Qi, X. Gao, Z. Luo and K. Cen, Environ Sci Technol, 2013, 47, 10056–10062.). Copyright (2013) American Chemical Society.

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Catal. Sci. Technol., 2015, 5, 3459–3472 | 3463

remove NO_x because of its higher efficiency, selectivity and economic feasibility. Generally, typical commercial SCR catalysts are composed of TiO_2, the catalytically active component V_2O_5, WO_3 and/or MoO_3 as promoter. The vanadia phase V_2O_5 not only catalyzes NO_x reduction but also catalyzes Hg^0 oxidation. Stolle et al.[85] observed that Hg^0 oxidation activity increased with increasing V_2O_5 concentration on SCR-DeNO$_x$-catalyst (Fig. 5). The highest Hg^0 oxidation activity was measured to be 86.6 m h^{-1} on the H7 catalyst with 2.6 w.t.% V_2O_5, while the lowest oxidation factor was measured to be 8.2 m h^{-1} on the almost vanadium-free test catalyst H8. This was well consistent with the previously reported literature[86] wherein Hg^0 oxidation increases almost linearly with VO_x loadings up to 10 w.t.%. WO_3 inhibits the initial sintering of TiO_2 and improves SO_2 resistance. On the other hand, WO_3 increases the amounts of Lewis acid. For V_2O_5-MoO_3/TiO_2 catalysts, Hg^0 oxidation was found to follow the Mars–Maessen mechanism. MoO_3 could not directly oxidize Hg^0, but molybdenum in high valence could assist to oxidize vanadium in low valence to increase the number of lattice oxygen for mercury oxidation.[2]

The SCR system is effective in controlling NO_x emissions as well as oxidizing Hg^0 in coal-fired flue gas. Nevertheless, it has some disadvantages, such as narrow active temperature window and toxicity to environment and human health due to the loss of vanadium during the preparation and operation processes.[87] Moreover, the conventional SCR catalysts were not effective enough for Hg^0 oxidation with low HCl concentrations. In addition, SO_2 and NH_3 have been observed to inhibit the oxidation of Hg^0 over the conventional SCR catalysts.[65] Hence, in order to overcome these disadvantages, many efforts have been paid to modify the catalysts. RuO_2 has been studied for modifying conventional SCR catalysts.[88] RuO_2 not only showed rather high catalytic activity on Hg^0 oxidation by itself, but also appeared to be well cooperative with the commercial SCR catalyst for Hg^0 conversion. Besides, the modified commercial SCR catalyst with RuO_2 displayed an excellent tolerance to SO_2 and NH_3 without any distinct negative effects on NO_x reduction and SO_2

Fig. 5 Mercury oxidation activity f_{Hg} of commercial SCR DeNO$_x$ catalysts as function of their V content (influent model flue gases with 40 μg m^{-3} elemental mercury, 100 mg m^{-3} HCl, 4% by volume (dry) O_2, 7% by volume H_2O and no NO, NH_3 and SO_2, 390 °C).[85]
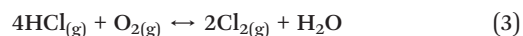
conversion. At the same time, the Hg^0 oxidation activity of commercial SCR catalysts impregnated with different metal oxides (Cr_2O_3, ZnO, CuO, NiO, MnO) was also investigated.[89] Results showed that the Hg^0 oxidation efficiency of metal oxide-impregnated commercial SCR catalysts was higher than that of the non-impregnated reference commercial SCR catalyst. Especially, CuO/SCR catalyst exhibited the best Hg^0 oxidation activity.

4. Proposed mechanism for the catalytic oxidation of elemental mercury

Hg adsorption and oxidation on catalyst surfaces has been studied in a number of research.[64,90–92] It is well known that Hg^0 can undergo either heterogeneous or homogeneous reactions on the SCR systems. SCR catalysts are believed to facilitate heterogeneous oxidation, which has a faster reaction rate than homogeneous oxidation.[93] However, the exact mechanisms for Hg oxidation on SCR catalysts and their dependence on flue gas properties were not yet well understood. In order to obtain an understanding of the mechanisms governing Hg^0 oxidation, the effect of SCR catalysts for Hg^0 oxidation has been widely studied.[90,91,94] Several mechanisms, including the Deacon process, the Eley–Rideal mechanism, the Langmuir–Hinshelwood mechanism and the Mars–Maessen mechanism, have been used to explain the heterogeneous Hg^0 oxidation.

4.1 Deacon reaction

The Deacon process[95] generates Cl_2 by catalytic oxidation of HCl with air or oxygen, which takes place at about 300–400 °C (eqn (3)).

$$4HCl_{(g)} + O_{2(g)} \leftrightarrow 2Cl_{2(g)} + H_2O \qquad (3)$$

In the presence of an appropriate catalyst, the Deacon process could convert large concentrations of HCl into Cl_2, and the generated Cl_2 is the key factor for Hg oxidation in the flue gas. Copper, iron, and manganese salts are suitable catalysts for the Deacon process. Hisham and Benson et al.[96] studied the basic thermochemistry of the Deacon reaction over a large number of groups and transition metal oxides. The process comprises a catalytic cycle which can be examined in terms of two independent steps: (1) HCl absorption by the metal oxide to form the metal chloride (or oxychloride) plus water and (2) oxidation of chloride by O_2 to regenerate the metal oxide and free Cl_2. However, the Deacon reaction was described by a Mars–van Krevelen type mechanism involving five steps: hydrogen abstraction from HCl, recombination of atomic chlorine, hydroxyl recombination, water desorption and dissociative oxygen adsorption.[97] By invoking the Deacon mechanism, Du[98] suggested that $Cu_2Cl(OH)_3$ formed on the surface of absorbents and decomposed to CuCl in the
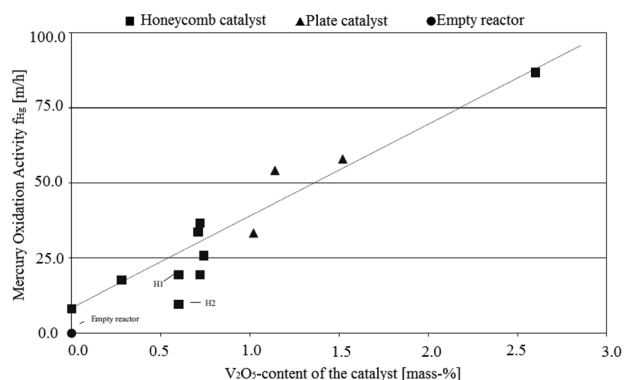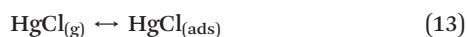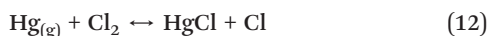
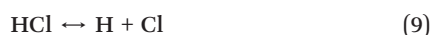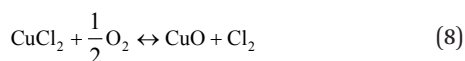3464 | Catal. Sci. Technol., 2015, 5, 3459–3472

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reaction of Hg removal, and then CuCl reacts with HCl (or O_2) to form little Cl_2. The mechanism is described as follows:

$$2Cu_2Cl(OH)_3 \leftrightarrow 2CuCl + 2CuO + 3H_2O + \frac{1}{2}O_2 \tag{4}$$

$$2CuCl + \frac{1}{2}O_2 \leftrightarrow Cu_2OCl_2 \tag{5}$$

$$Cu_2OCl_2 + 2HCl \leftrightarrow 2CuCl_2 + 2H_2O \tag{6}$$

$$CuO + 2HCl \leftrightarrow CuCl_2 + H_2O \tag{7}$$

$$CuCl_2 + \frac{1}{2}O_2 \leftrightarrow CuO + Cl_2 \tag{8}$$

$$HCl \leftrightarrow H + Cl \tag{9}$$

$$Cl_2 \leftrightarrow Cl + Cl \tag{10}$$

$$Hg + Cl \leftrightarrow HgCl \tag{11}$$

$$Hg_{(g)} + Cl_2 \leftrightarrow HgCl + Cl \tag{12}$$

$$HgCl_{(g)} \leftrightarrow HgCl_{(ads)} \tag{13}$$

$$HgCl_{2(g)} \leftrightarrow HgCl_{2(ads)} \tag{14}$$

$$HgCl_{(ads)} + Cl \leftrightarrow HgCl_{2(ads)} \tag{15}$$

4.2 Eley–Rideal mechanism

Senior and Linjewile[99] proposed that the mercury oxidation could occur *via* an Eley–Rideal mechanism. HCl competes with NH_3 for surface active sites, and adsorbed HCl reacts with gaseous (or as a weakly adsorbed) Hg^0 (eqn (16)–(17)).

$$HCl_{(g)} \leftrightarrow HCl_{(ads)} \tag{16}$$

$$HCl_{(ads)} + Hg^0_{(g)} \rightarrow HgCl_{2(g)} \tag{17}$$

On the other hand, Senior[100] suggested the Eley–Rideal type mechanism in which Hg^0 adsorption was in competition with NH_3 adsorption and adsorbed Hg^0 reacts with gaseous HCl. Recently the V_2O_5-based SCR catalysts were found to oxidize Hg^0 to Hg^{2+}, which might follow the Eley–Rideal mechanism.[65,101] According to this mechanism, HCl is dissociatively adsorbed on V_2O_5-active sites. Then the chemically adsorbed Cl species reacts with gas-phase Hg^0 to generate an intermediate HgCl species, which further reacts with chlorine species to form $HgCl_2$. Interestingly, the reaction between Hg^0 and H_2S was in a similar manner, whereby active surface sulfur reacts with gas phase Hg^0 to form stable HgS.[83] The possible reactions are proposed as follows:

$$H_2S + O^* \rightarrow S_{(ads)} + H_2O \tag{18}$$

$$S_{(ads)} + Hg \rightarrow HgS \tag{19}$$
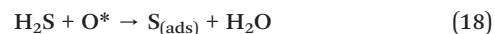
where $S_{(ads)}$ and O^* are active surface sulphur and surface oxygen of the sorbent, respectively.

4.3 Langmuir–Hinshelwood mechanism

The bimolecular reaction between two species adsorbed on a surface can be described by a Langmuir–Hinshelwood mechanism.[102] Wang *et al.*[103] suggested that mercury oxidation on the MnCe catalyst surface followed the Langmuir–Hinshelwood mechanism, where reactions took place between the adsorbed active species and adsorbed Hg^0 to form Hg^{2+}. It has also been reported that Hg^0 oxidation over V_2O_5-based catalyst occurred *via* the Langmuir–Hinshelwood mechanism. On the basis of this mechanism, gas-phase Hg^0 and HCl adsorbed onto the vanadia sites to form $HgCl_2$ and V–OH species. Then, the reoxidation of the V–OH species by oxygen follows to form V=O and H_2O.[66] The possible pathway responsible for mercury oxidation over V_2O_5-based catalyst is described in Fig. 6. A similar mercury oxidation mechanism over Ce–Ti catalyst was proposed by other authors.[84,104] Specifically, in Suarez Negreira's studies,[105,106] it was showed that Hg^0 had a negligible interaction with the vanadia oxide dimer, while HgCl had the strongest adsorption, followed by HCl. The proposed Hg^0 oxidation mechanism may contain the following two steps: first, Langmuir–Hinshelwood step between HCl and HgCl to produce $HgCl_2$ and second, Eley–Rideal step between gas-phase Hg^0 and adsorbed HCl to produce HgCl. Based on these results, the mechanism of Hg^0 oxidation through the formation of $HgCl_2$ is proposed in Fig. 7.
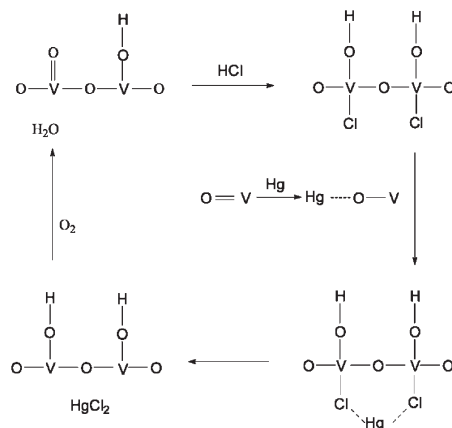

Fig. 6 Mechanism of the mercury oxidation on the vanadia-based SCR catalysts.[66] Reprinted (adapted) with permission from (S. He, J. S. Zhou, Y. Q. Zhu, Z. Y. Luo, M. J. Ni and K. F. Cen, Energy Fuels, 2009, 23, 253–259.). Copyright (2009) American Chemical Society.
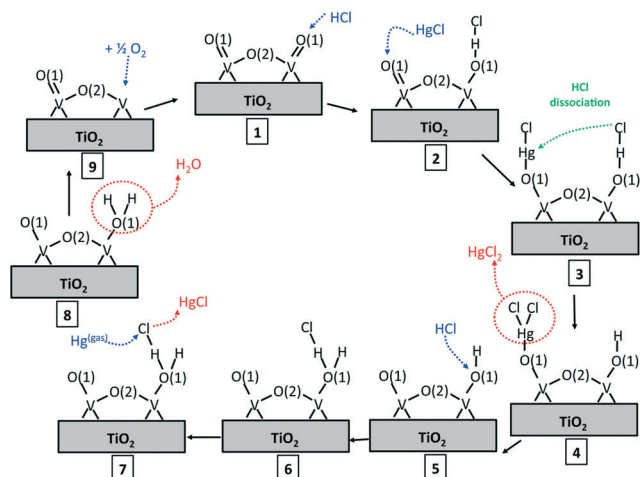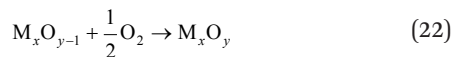
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Catal. Sci. Technol., 2015, **5**, 3459–3472 | **3465**

Fig. 7 Proposed mechanism of mercury oxidation on vanadia-titania SCR catalyst. Blue arrows indicate an adsorption step, green arrows indicate a dissociation step, and red arrows indicate a desorption step.[105] Reprinted (adapted) with permission from (A. Suarez Negreira and J. Wilcox, J. Phys. Chem. C, 2013, 117, 1761–1772.). Copyright (2013) American Chemical Society.

4.4 Mars–Maessen mechanism

Initially, Zhang et al.[107] proposed that Hg^0 oxidation on Co_xMn_yTi catalyst can be interpreted by the Mars–Maessen mechanism, where Hg^0 bonds with lattice oxygen and/or chemisorbed oxygen of the catalyst surface to form weakly bonded speciation $Hg–O–M–O_{x-1}$ (M = Mn or/and Co) and then formed mercuric oxide (HgO). The consumed lattice oxygen and/or surface oxygen can be replenished by the gas-phase O_2. The Mars–Maessen mechanism has been widely used for illustrating the Hg^0 oxidation process on metal oxide catalysts.[68,108–111] In this mechanism, adsorbed Hg^0 would react with a lattice oxidant (either O or Cl) that is replenished from the gas phase, forming a binary mercury oxide.[59] Reaction eqn (20)–(24) showed the Mars–Maessen mechanism for the reaction of adsorbed Hg^0 with lattice oxidant.

$$Hg_{(g)} \rightarrow Hg_{(ads)} \tag{20}$$

$$Hg_{(ads)} + M_xO_y \rightarrow HgO_{(ads)} + M_xO_{y-1} \tag{21}$$

$$M_xO_{y-1} + \frac{1}{2}O_2 \rightarrow M_xO_y \tag{22}$$

$$HgO_{(ads)} \rightarrow HgO_{(g)} \tag{23}$$

$$HgO_{(ads)} + M_xO_y \rightarrow HgM_xO_{y+1} \tag{24}$$

5 Kinetics

It is known that Hg conversions in flue gas are kinetically, but not thermodynamically, controlled.[112] A kinetic model is a useful tool to simulate the mercury oxidation and evaluate the mercury oxidation efficiency for various operational conditions. It can hopefully facilitate the predictions of the effectiveness of different mercury control measures and strategies. Lots of studies had been done for researching the mechanisms and kinetics of Hg^0 oxidation.

As tabulated in Table 3, formulation of a reaction mechanism began with the kinetic framework of Widmer.[113] The reaction controlling the Hg conversion is between Hg^0 and the chlorine atom.[114] Although such mechanisms can give plausible qualitative results, they are generally not suitable for examining the effects of other flue gas constituents on Hg chlorination.[45] It should be recognized that Hg oxidation would be subjected to kinetic control under actual combustion conditions. As a result, it is necessary to develop mercury chemical kinetics for application in real combustion systems.[114] Presto et al.[115] researched a kinetic approach to the catalytic oxidation of Hg^0 in flue gas. They proposed a method for analysing Hg^0 oxidation catalyst results in a kinetic framework using the bulk reaction rate for oxidized mercury formation normalized by either the catalyst mass or surface area. The bulk Hg^0 oxidation reaction rate (R_{cat}) can be described as an apparent gas-phase reaction as shown in eqn (25):

$$R_{cat} = R_{gas}\left(\frac{V_{cat}}{m_{cat}}\right) = k_{cat}\left[Hg^0\right]^\alpha \left[Oxidant\right]^\beta \tag{25}$$

where R_{gas} is the bulk reaction rate for Hg^{2+} formation across the catalyst bed, V_{cat} is the catalyst volume, m_{cat} is the catalyst mass, k_{cat} is the catalyst-normalized rate constant. Results reported for fractional mercury oxidation are strongly influenced by the specific experimental conditions and therefore difficult to translate from experiment to experiment.

Li et al.[116] developed and evaluated a kinetic modelling of Hg^0 oxidation by chlorine over CeO_2-TiO_2 catalysts. Based on the conditions of the experiment and assuming that Hg^0 oxidation over CeTi catalysts follows the Langmuir–Hinshelwood mechanism, the bulk Hg^0 oxidation reaction rate under flue gas containing HCl can be described as:

$$R_{cat} = -k\left[Hg^0\right]^S \theta_{HCl} = -k\left[Hg^0\right]^S \frac{k_{HCl}\left[HCl\right]}{1 + k_{HCl}\left[HCl\right]} \tag{26}$$

where K is the overall reaction rate constant, $[Hg^0]^s$ is the concentration of surface-phase Hg^0, θ_{HCl} is the fraction of the

Table 3 Rate constants in Hg^0 oxidation mechanism[113]

No	Reactions	A cm³ mol⁻¹ s⁻¹	β	E_α kcal mol⁻¹
1	$Hg + Cl + M = HgCl + M$	2.40×10^8	1.4	−14.4
2	$Hg + Cl_2 = HgCl + Cl$	1.39×10^{14}	0.0	34.0
3	$HgCl + Cl_2 = HgCl_2 + Cl$	1.39×10^{14}	0.0	1.0
4	$HgCl + Cl + M = HgCl_2 + M$	2.19×10^{18}	0.0	3.10
5	$Hg + HOCl = HgCl + OH$	4.27×10^{13}	0.0	19.0
6	$Hg + HCl = HgCl + H$	4.94×10^{14}	0.0	79.3
7	$HgCl + HCl = HgCl_2 + H$	4.94×10^{14}	0.0	21.5
8	$HgCl + HOCl = HgCl_2 + OH$	4.27×10^{13}	0.0	1.0

3466 | Catal. Sci. Technol., 2015, 5, 3459–3472

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active sites occupied by HCl, k_{HCl} is the Langmuir adsorption constant of HCl, and [HCl] is the gas-phase HCl concentration. Experimental data were analyzed using a kinetic model incorporating mass transfer, adsorption equilibrium and mass balance with key variables of interest being residence time, catalyst type and HCl concentration in simulated coal combustion flue gas. The experimental data verification for the proposed kinetic model was satisfactory, indicating the validity of the model for describing the mechanism of Hg^0 oxidation by chlorine over CeTi catalysts.

The reaction mechanisms and catalytic oxidation kinetics in the oxidizing flue gas were also evaluated by Gao et al.[117] In their study, the kinetic model following the Eley–Rideal mechanism was implemented. The reaction rate of Hg^0 oxidation (r) defined in eqn (27) can be expressed as the change in reactant concentration:

$$\gamma = k_2 C_{Hg^0}^x C_{O_2}^y \gamma_1 = k_2 C_{Hg^0}^x C_{O_2}^y \left(-F_{HCl}^0 \frac{d\eta_{HCl}}{dw} \right) = K^* C_{Hg^0}^x C_{O_2}^y \quad (27)$$

where k^* is the overall rate constant for oxidation, k_2 is the reaction rate constant for Hg^0, and x and y are the reaction order with respect to Hg^0 and O_2, respectively. The results showed that the rate of Hg^0 oxidation over commercial SCR catalyst was in zeroth order with respect to O_2 and nearly first order with respect to Hg^0. In agreement with the experimental results, the kinetic model well described the rate of Hg^0 oxidation in the presence of HCl and O_2 in different conditions. According to the kinetic model, the apparent activation energy for Hg^0 oxidation over the commercial SCR catalyst was 37.73 kJ mol^{-1}.

6 Effect of flue gas constituents (HCl, H_2O, SO_2, O_2, NO, NH_3) on Hg^0 oxidation

Hg^0 oxidation efficiency highly depended on the composition of flue gas. Although the SCR catalyst is active for Hg^0 oxidation, its effectiveness was found to depend on the halide species and their concentration.[118] Many studies suggested that HCl plays the most important role in oxidation of Hg^0.[74,119–121] It could remarkably increase Hg^0 oxidation. He et al.[66] analyzed the effect of HCl on the Hg^0 oxidation across the SCR unit, as shown in Fig. 8. The results indicated that the monomeric vanadyl sites on the catalyst surface were found to be responsible for the adsorption of both Hg^0 and HCl, which meant that they were active for mercury oxidation. The detailed Langmuir–Hinshelwood mechanism was proposed to explain the Hg oxidation on vanadia-based SCR catalyst, where reactive Cl generated from adsorbed HCl reacts with adjacent Hg^0. At the same time, it has been recognized that the oxidation of Hg^0 over catalysts in the presence of HCl might experience a series of reactions, such as Deacon reaction, Mars–Maessen or Eley–Rideal mechanism.[11,65,122,123] Meanwhile, the reaction of HCl and the active component
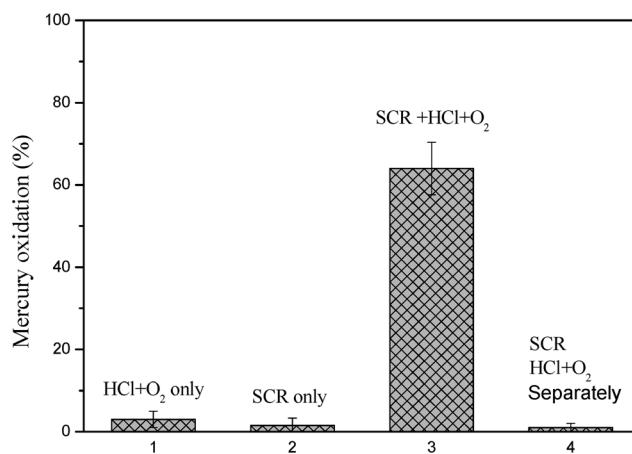

Fig. 8 The effect of HCl on the heterogeneous oxidation of Hg across the SCR unit.[66] Reprinted (adapted) with permission from (S. He, J. S. Zhou, Y. Q. Zhu, Z. Y. Luo, M. J. Ni and K. F. Cen, Energy Fuels, 2009, 23, 253–259.). Copyright (2009) American Chemical Society.

V_2O_5 of the SCR catalysts were also investigated. For example, Liu et al.[124] studied the HCl adsorption on different sites of V_2O_5 (0 0 1) surface, and the comparison between HCl and Hg adsorptions was made. The results showed that Hg^0 adsorption on the V_2O_5 surface is stronger than that of HCl. The Hg^0 in the flue gas is adsorbed by the V_2O_5 surface as an intermediate which reacts fairly rapidly with chlorine species to form surface HgCl, and then reacts with chlorine species to form surface $HgCl_2$. Finally, $HgCl_2$ desorbs from the V_2O_5 surface.

It should be noted that HBr also shows a very strong impact on increasing Hg^0 oxidation.[125] Results by Cao et al.[126] suggest that the promotional effect of HBr is far more pronounced than that of HCl. On cooling of the gases, the diatomic and molecular forms of the halogens become stable according to the Deacon type of reactions:[41,127]

$$4HCl + O_2 \leftrightarrow 2H_2O + 2Cl_2 \quad (28)$$

$$4HBr + O_2 \leftrightarrow 2H_2O + 2Br_2 \quad (29)$$
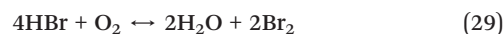
However, the depletion of Cl_2 would occur by the enriched SO_2 in the coal-derived flue gases.[127] Br_2 oxidizes the typical amounts of mercury in the coal flue gases through direct mercury bromination. This explains why HBr is a more effective mercury oxidizer than HCl.

$$SO_2 + Cl_2 + H_2O \rightarrow SO_3 + 2HCl \quad (30)$$
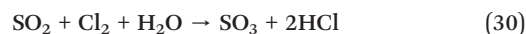
H_2O has been reported to inhibit Hg^0 oxidation over metal oxide catalysts due to competitive adsorption.[101,128] The competitive adsorption of water vapor on the active site may have prohibited the adsorption of reactive species that have a promotional effect on Hg^0 oxidation such as O_2, HCl and NO_x.[129–131] For instance, the conversion of Hg^0 to Hg^{2+} may decrease due to the competition between the O and OH when both water vapor and oxygen co-exist in the simulated flue

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Catal. Sci. Technol., 2015, 5, 3459–3472 | 3467

gas. Furthermore, a high concentration of water vapor in the flue gas would diminish the adsorption of HCl, which is a crucial flue gas component responsible for Hg^0 oxidation.

The effect of SO_2 on Hg^0 oxidation in flue gas was not conclusive, either promotional or inhibitory.[49,132] Wan et al.[82] would like to define SO_2 as a promoter because the addition of 800 ppm SO_2 slightly enhanced the Hg conversion under dry conditions. It might be supposed that the sulfate species could be more easily formed since SO_2 was introduced into the reaction. The formed sulfate species adsorbed on the catalyst surface could also enhance the catalytic activation because of the newly formed sulfate adsorption acid sites.[133] On the contrary, the inhibitory effect of SO_2 on Hg^0 oxidation had been shown in many reports.[61,103,121,134,135] On the one hand, SO_2 had a large inhibitory effect on Hg^0 oxidation mainly via the elementary reactions and to eliminate OH radicals.[135] On the other hand, SO_2 negatively affects Hg^0 oxidation due to the competitive adsorption between SO_2 and Hg^0 on active sites.[61,121] In particular, Li et al.[84,136] showed that SO_2 had different effects on Hg^0 oxidation under different flue gas conditions. SO_2 inhibited Hg^0 oxidation without O_2, while SO_2 had a promotional effect on Hg^0 oxidation in the presence of O_2. As shown in Fig. 9, without O_2, addition of SO_2 into gas flow containing 300 ppm NO decreased Hg^0 oxidation efficiency from 38.3% to about 6.5%. With the aid of 4% O_2, the addition of 400 ppm SO_2 into gas flow containing 300 ppm NO increased Hg^0 oxidation efficiency from 45.6% to 64.7%. Conversion of SO_2 to SO_3 not only relieved the prohibitive effect related to the competitive adsorption between SO_2 and Hg^0 but also yielded active SO_3 for Hg^0 oxidation, and hence has an obvious promotional effect.

O_2 plays a crucial role in Hg^0 oxidation.[137] To obtain higher Hg^0 oxidation efficiency, the presence of O_2 is normally necessary, especially for metal oxide catalysts.[68,69] Some researchers believed that Hg^0 oxidation in the presence of O_2 can be explained by the Mars–Maessen mechanism.[107] Hg^0 reacts with the lattice oxygen on the catalyst surface to form weakly bonded species and then formed HgO. The consumed lattice oxygen can be replenished by the gaseous O_2. Yet researchers believed that adsorbed O_2 could oxidize Hg^0 to HgO directly.[138] However, Xu et al.[45] found that O_2 showed

little effect on Hg^0 oxidation alone. It should be noted that O_2 has a synergistic effect on mercury oxidation via SO_2, NO or HCl.[84,130,139]

NO can achieve a significant promotional effect on Hg^0 oxidation. It has been reported that adsorbed NO can be oxidized by the surface oxygen of catalysts to give rise to species like NO^+, NO_2, nitrite, and nitrate, which are likely responsible for Hg^0 oxidation.[101,134] In contrast, the effect of NO on Hg^0 oxidation over the Mn–Ce/Ti catalyst was found to be inhibitory.[140] In the absence of O_2, NO is weakly adsorbed on the MnO_x-CeO_2 mixed oxides,[141] and a fraction of it reacts with the surface oxygen to form limited NO_2, nitrite, and nitrate species.[77] It was hypothesized that NO covered the active sites and consumed surface oxygen. Therefore, NO could cause a significant deteriorate effect on Hg^0 oxidation without the aid of O_2. Nevertheless, more adsorbed NO can be oxidized on the Mn–Ce/Ti catalyst to form abundant active species like NO_2 in the presence of O_2, which are more active than NO for Hg^0 oxidation.

The effect of NH_3 on Hg^0 oxidation has been analyzed several times with different results. A large number of studies suggested that NH_3 could cause a significant deteriorate effect on Hg^0 oxidation by competing for active sites with Hg^0.[142,143] However, it was pointed out that the oxidation rate of Hg^0 at the outlet of the SCR unit is not influenced by the injection of stoichiometric amounts of NO and NH_3.[144] Recently some studies have reported that Hg^{2+} is reduced by NH_3.[79,146] Thorwarth et al.[145] found that not only does Hg^0 oxidation not occur when NH_3 is present, but also NH_3 can reduce Hg^{2+} to Hg^0. At temperatures higher than 325 °C, NH_3 may also reduce Hg^{2+} in accordance with the following reaction:[146]

$$3HgCl_2 + 2NH_3 \rightarrow 3Hg_{(g)} + N_2 + 6HCl \qquad (31)$$
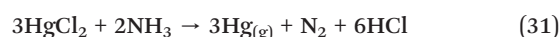
In addition to this effect, the $DeNO_x$ reaction may also cause a reduction of Hg^{2+}.[85] It is assumed that the reduction of the Hg^{2+} is caused by the intermediate reduced vanadium species produced by the $DeNO_x$ reaction in the catalysts. The sum reaction involving $DeNO_x$ and mercury could be formulated as:
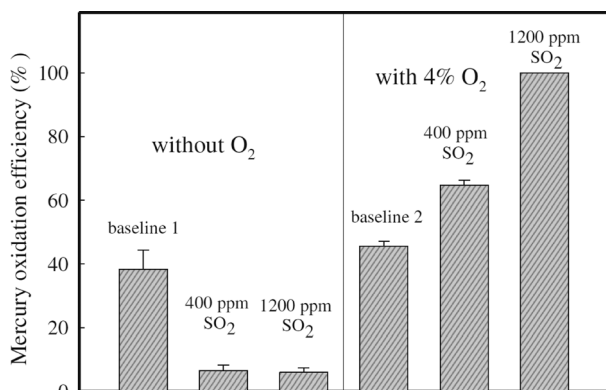
$$6NO + 6NH_3 + 3HgCl \rightarrow 6N_2 + 3Hg_{(g)} + 6H_2O \qquad (32)$$

7 Effect of space velocity and temperature

Some studies suggest that increasing space velocity reduces Hg^0 oxidation activity across the SCR unit. For instance, Machalek et al.[147] observed that the extent of Hg^0 oxidation was reduced from 40% to 5% as the gas space velocity increased from 3000 to 7800 h^{-1} in flue gases derived from subbituminous coal. Another study by Lee et al.[148] reported that Hg^0 oxidation decreased from 88 to 53% as space velocity increased from 2000 to 4000 h^{-1} at 350 °C.

Fig. 9   Effect of SO_2 on Hg^0 oxidation in the presence of NO.[136]

3468 | Catal. Sci. Technol., 2015, 5, 3459–3472

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Reaction temperature has a strong influence on Hg^0 oxidation. The efficiency of Hg^0 oxidation increased with increasing temperature in SCR conditions.[149] However, some studies suggest that Hg^0 oxidation decreased with increasing temperature. Rallo *et al.*[142] observed a decrease in Hg^0 oxidation from 70% at 280 °C to 50% at 350 °C.

8 Conclusions and outlook

This work focused on catalytic oxidation of elemental mercury by SCR catalysts in coal-fired flue gas. Mercury is a global pollutant which is emitted into the atmosphere from natural and various anthropogenic sources. Coal-fired power plants are considered to be a major source of mercury emissions from anthropogenic activities. Mercury transformations and speciation in combustion flue have been studied intensively. Hg in coal was found to be volatile. Most of them entering the furnace were rapidly volatilized and present in the gas phase. In the post combustion section, Hg reacts with flue gas constituents to convert to gaseous oxidized forms and particulate-bound mercury. Eventually, the principal forms of Hg in coal combustion flue gas are assumed to be Hg^0, Hg^{2+}, and Hg^p.

In addition to NO_x control, SCR catalysts affect the Hg speciation by altering Hg^0 to Hg^{2+}. Increasing the emission of Hg^{2+} across the SCR catalyst allows for high reduction of Hg emission because Hg^{2+} can be removed in downstream equipment such as WFGD systems. A review on the recent advances in catalytic oxidation of Hg^0 by SCR catalysts in flue gas was provided. SCR catalysts including noble metal and non-noble metal catalysts have been summarized. Among different noble metals, Pd and Au are the most attractive options for controlling Hg^0 emissions. Several non-noble metals including MnO_2, CeO_2, V_2O_5, *etc.* have also showed impressive Hg^0 oxidation activity. One unresolved problem is how to improve their SO_2 resistance and Hg^0 oxidation efficiency simultaneously. Meanwhile, the influence of flue gas constituents, space velocity and temperature is very significant.

Different mercury reaction mechanisms including Deacon mechanism, Eley–Rideal mechanism, Langmuir–Hinshelwood mechanism, and Mars–Maessen mechanism were also proposed to describe Hg^0 oxidation in flue gas. At the same time, various kinetic models have been developed to simulate mercury oxidation. Although the assumptions were very different, the experimental data verification for the proposed kinetic model was satisfactory.

The catalytic mercury oxidation in flue gases is an actively researched area. But there are still many challenges, such as the conventional SCR catalysts were not effective enough for Hg^0 oxidation in the absence of HCl. What is more, mercury oxidation under an SCR atmosphere was not effective enough because of the inhibitory effect of ammonia. It should be noted that the conversion of SO_2 to SO_3 over SCR catalysts should not improve when promoting Hg^0 oxidation activity. Furthermore, the exact mechanisms for Hg^0 oxidation by SCR catalysts and their dependence on flue gas properties were not yet well understood. Therefore, further investigation is necessary for developing new and efficient SCR catalysts with a low SO_2/SO_3 conversion rate. The role of flue gas components and the mechanism of Hg^0 oxidation need to be better understood as well.

Acknowledgements

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