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REVIEW

Recent developments on gas–solid heterogeneous oxidation removal of elemental mercury from flue gas

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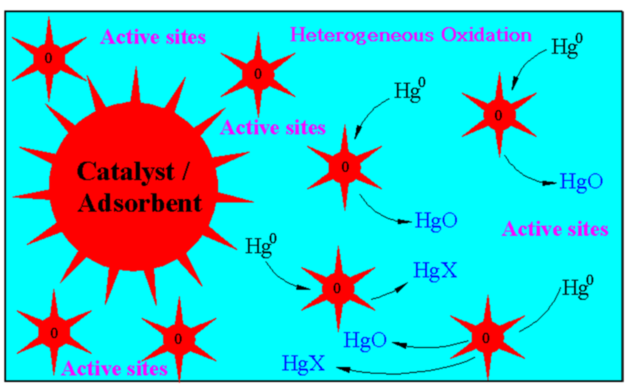
Abstract

Mercury is a toxic and persistent environmental pollutant which has been recognized as a global threat to human health and our ecosystem because mercury bio-accumulates in the food chain and can be transformed into the more neurotoxic methylmercury. Among current and emerging abatement technologies for elemental mercury in flue gas, gas–solid heterogeneous oxidation is nowadays gaining increasing attention due to several inherent advantages. The catalysts and adsorbents are key materials that control the heterogeneous catalytic oxidation and adsorption of Hg^0 from flue gas. Here we present a review of the recent developments on several catalysts and adsorbents, including noble metal-based catalysts, non-noble metal-based catalysts (transition metal oxides and selective catalytic reduction catalysts), activated carbon/coke-based sorbents, biochar-based sorbents, fly ash-based sorbents, mineral material-based sorbents and other novel catalysts. The key process parameters and kinetic reaction mechanisms and advantages and disadvantages of various emerging catalysts/adsorbents and technologies of Hg^0 removal are described in detail.

Graphical abstract



Keywords Flue gas · Hg^0 removal · Gas–solid heterogeneous oxidation · Sorbents or catalysts

Introduction

Mercury is an environmental persistent pollutant of great public concern because of its well-known high neurological toxicity, and well-documented food chain transport and bioaccumulation in its different forms, such as methylmercury with its concomitant adverse effects on our ecosystem and human health (Li et al. 2009). Human exposure to mercury occurs primarily by consumption of contaminated

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fish, resulting in such detrimental effects on human health, including neurological disorders, kidney damage, and birth defects. Therefore, United States Environmental Protection Agency (US EPA) identified mercury as a toxic and hazardous air pollutant under Title III of the 1990 Clean Air Act Amendments (CAAA) (Qiao et al. 2009). The total amount of anthropogenic mercury emission is about 1000–6000 tons per year (Yang et al. 2007). Combustion activities such as the burning of fossil fuels, municipal solid wastes, and medical wastes are the largest source of mercury emissions, which accounts for more than 90% of all anthropogenic mercury emissions (Reddy et al. 2012). To abate mercury emissions, some countries and regions have developed very stringent laws. In 2011, the United States Environmental Protection Agency (US EPA) promulgated the first national standard for mercury emissions, namely the Mercury and Air Toxics Standards (MATS), which aims to limit emissions of mercury and other toxic substances in power plants (Gao et al. 2013b). Also, in 2013, the United States Environmental Protection Agency updated the national emission standard (MATS), stipulating mercury emission limit below 0.003 lb GWh^{-1} (Zhao et al. 2015b). In July 2011, the State Environmental Protection Administration of China (SEPA) released a new national standard (GB 13223-2011) of air pollutants for power plants, which requires new coal-burning power plants' atmospheric mercury emissions should be less than 30 $\mu g/m^3$ (Ancora et al. 2016). Therefore, the need and knowhow to curb mercury emissions are nowadays gaining significant global attention.

To reduce the emission of air pollutants, most coal-fired power plants have been installed some air pollution control devices (APCDs). Fabric filters (FF) and electrostatic precipitators (ESP), wet flue gas desulfurization (WFGD) system, and selective catalytic reduction (SCR) devices can effectively control particulate matter, SO_2 and NO_x in flue gas, respectively. During the combustion process, the elemental mercury in fuel is released into the flue gas in the form of vapor. This gaseous elemental mercury is subsequently oxidized partially to Hg^{2+} by heterogeneous (gas–solid) and homogeneous (gas–gas) reactions (Lee et al. 2002). Therefore, mercury in typical flue gas consists of three forms: elemental mercury (Hg^0), oxidized mercury (Hg^{2+}) and particulate-bound mercury (Hg^p) (Chi et al. 2009). Some studies have reported that the existing conventional air pollution control devices (APCDs) for reducing emissions of SO_2, NO_x and particulate matter can achieve a certain degree of mercury removal (Zheng et al. 2011; Wang et al. 2010b). For example, the Hg^{2+} can be efficiently removed by the existing wet flue gas desulfurization (WFGD) equipment due to its high water solubility (Li et al. 2011a). Fabric filters (FF) and/or electrostatic precipitators (ESP) can easily capture Hg^p from flue gas (Cao et al. 2008). In contrast, Hg^0 with high volatility and low solubility in water is very difficult to be effectively removed by existing APCDs (Gutiérrez et al. 2007; Galbreath and Zygarlicke 1996). To reduce operating costs, the use of existing conventional APCDs to remove elemental mercury from flue gas is considered as an effective option for mercury abatement. Therefore, one of the core issues of mercury emission control is the efficient oxidation of elemental mercury (Hg^0) into the oxidized form (Hg^{2+}).

To effectively control mercury emissions from coal-fired boilers, many Hg^0 control technologies have been developed over the past few decades, including adsorptive removal (Vidic and Siler 2001; Tan et al. 2012a; Chung et al. 2009), catalytic oxidation (Wang et al. 2010a; He et al. 2013), advanced oxidation (Wang et al. 2010c; An et al. 2014; Xu et al. 2008), and traditional chemical oxidation technologies (Wang et al. 2007; Hutson et al. 2008; Stergarsek et al. 2010). Adsorption processes utilizing modified and supported sorbents can effectively remove Hg^0 in flue gas by converting it to Hg^p and Hg^{2+} (Pavlish et al. 2004; Wu et al. 2012). In addition, catalytic oxidation processes such as selective catalytic reduction (SCR) and using catalysts composed of noble metals, metal oxides, and multi-metal oxides can simply, efficiently, and cost-effectively oxidize Hg^0 to Hg^{2+} (Kamata et al. 2009; Yang et al. 2010). Among technologies for Hg^0 removal from flue gas, the gas–solid heterogeneous adsorption and catalytic oxidation are recognized as the most promising (Pavlish et al. 2004; Wu et al. 2012; Kamata et al. 2009; Yang et al. 2010). While some reviews on mercury control have been published in the past few decades, these reviews appear to be limited in scope and/or outdated due to the prolific research productivity in this field, and hence, there is a need for a more comprehensive review of the recent developments and emerging technologies. (Gao et al. 2013b; Zheng et al. 2012; Pavlish et al. 2003; Fu et al. 2010). This exhaustive review discusses the emerging catalysts and adsorbents, including noble metal-based catalysts, non-noble metal-based catalysts (transition metal oxides and SCR catalysts), activated carbon- and coke-based sorbents, biochar-based sorbents, fly ash-based sorbents, mineral material-based sorbents, and other novel catalysts, in detail. Some challenges, problems, and future research directions of Hg^0 removal using these catalysts and adsorbents are also discussed. The key process parameters, advantages, and disadvantages of current and emerging technologies are summarized, and the reaction kinetics and mechanistic aspects of gas–solid heterogeneous catalytic oxidation and adsorption of Hg^0 from flue gas are described in detail.

Gas–solid heterogeneous oxidation of mercury

It is well known that Hg^0 in flue gas is very difficult to be captured due to its low solubility in water and high volatility. However, Hg^p can be removed in particle controllers, and the oxidized mercury (Hg^{2+}) can be easily captured by the wet flue gas desulfurization (WFGD) system due to its high water solubility. Therefore, a combination of wet flue gas desulfurization (WFGD) system and elemental mercury heterogeneous oxidation is considered as a promising method for Hg^0 control. To date, a number of heterogeneous catalyst and adsorption systems have been developed for Hg^0 oxidation or removal and categorized into seven groups, namely noble metal-based catalysts, non-noble metal-based catalysts, activated carbon-/coke-based sorbents, biomass char-based sorbents, fly ash-based sorbents, mineral material-based sorbents, and other novel catalysts.

Noble metal-based catalysts

Noble metals such as Au, Pd, Ag, Ru, and Ir have been considered as potential Hg^0 oxidation catalysts due to their regeneration performance and excellent mercury adsorption capacity. To obtain a high mercury removal capacity, the noble metals are usually supported on materials with well-developed pore structures and large Brunauer–Emmet–Teller (BET) surface areas, such as alumina, silica, zirconia, titania, carbons, and zeolite. The modification conditions and Hg^0 removal capacities of the investigated noble metal catalysts are summarized in Table 1.

Pd has been recognized as a promising catalyst for mercury removal (Granite et al. 2006). In the study by Hou et al. (2014a), the Pd catalyst exhibited high mercury removal efficiency in the operating temperature range of 200–270 °C, reporting that, up to 270 °C, a catalyst containing 8% Pd provided 90% Hg^0 removal efficiency and retained good stability at mid-temperatures. Li et al. (2014a) also tested the effects of three operating temperatures, 250, 300, 350 °C, on mercury adsorption capacity and found the mercury adsorption efficiency at 250 °C was higher than those at 300 and 350 °C, confirming the positive effects of operating at mid-temperatures. Hou et al. (2014a) and Han et al. (2012, 2016) tested the effects of H_2 and CO on mercury removal and observed that H_2 and CO could enhance the removal efficiency of elemental mercury as a result of the reduction of PdO to Pd metal. Hou et al. (2014a) also studied the effect of HCl on Hg^0 removal over Pd based catalyst and found that HCl promoted mercury removal. Yue et al. (2015) examined the effects of H_2S on mercury removal over Pd/AC catalyst and showed that H_2S significantly inhibited Hg^0

Table 1 Reaction conditions and Hg^0 removal performance of noble metal catalysts

Raw sorbents	Name of modified sorbents	Simulated flue gas	RT* (°C)	MRE**(%)	AC*** (μg/g)	References
Al_2O_3	Pd/Al_2O_3	$H_2/CO/H_2S/HCl/Hg^0$	270	> 90	–	Hou et al. (2014a)
Activated carbon (AC)	Pd/AC	$H_2/CO/H_2S/N_2/Hg^0$	250	94	4.84	Li et al. (2014a)
γ-Al_2O_3	1Pd3Fe/γ-Al_2O_3	N_2/Hg^0	250	> 80	–	Han et al. (2012)
Activated carbon (AC)	1Pd5Fe/AC	$N_2/H_2S/Hg^0$	200	> 80	–	Han et al. (2016)
	Pd/AC	$N_2/H_2S/H_2/Hg^0$	200	91.4	–	Yue et al. (2015)
Activated carbon (BAC)	BAC^{Cl-Au}	Air/Hg^0	–	> 97	10.0	Song and Lee (2016)
Carbon	Au/C	$SO_2/CO_2/N_2/Hg^0$	120	–	38.7	Gómez-Giménez et al. (2015)
Carbon	MC-Au-red	N_2/Hg^0	75	–	23	Ballestero et al. (2013)
TiO_2	Ag–Mo–TiO_2	$N_2/O_2/HCl/Hg^0$	150	> 90	–	Zhao et al. (2015d)
Selective catalytic reduction (SCR)	Ru-SCR	$N_2/O_2/HCl/Hg^0$	350	95	–	Chen et al. (2014)
TiO_2	RuO_2/TiO_2	$N_2/CO_2/O_2/SO_2/NO/NH_3/HCl/Hg^0$	350	> 90	–	Liu et al. (2016b)
		$N_2/CO_2/O_2/SO_2/NO/NH_3/HBr/Hg^0$		> 90		
Rutile TiO_2	RuO_2/rutile TiO_2	$N_2/CO_2/H_2O/O_2/NO/NH_3/HCl/Hg^0$	350	> 90	–	Liu et al. (2017)
$Ce_xZr_{1-x}O_2$	$IrO_2/Ce_xZr_{1-x}O_2$	$O_2/SO_2/HCl/N_2/Hg^0$	150	97	–	Chen et al. (2016)

*Reaction temperature

**Hg^0 removal efficiency

***Adsorption capacity

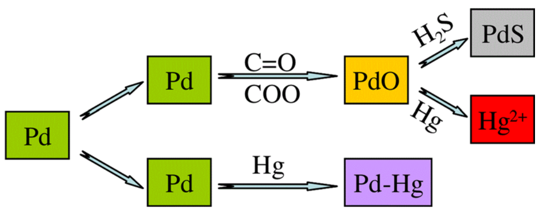

Fig. 1 Schematic diagram of the pathways of mercury removal over the Pd/activated carbon samples in N_2–Hg–H_2S atmosphere. The possible product in the N_2–Hg–H_2S atmosphere, PdS, is difficult to reduce to Pd^0, suggesting that this product could be inhibitory to the mercury removal process (reproduced with permission from Li et al. 2014a)

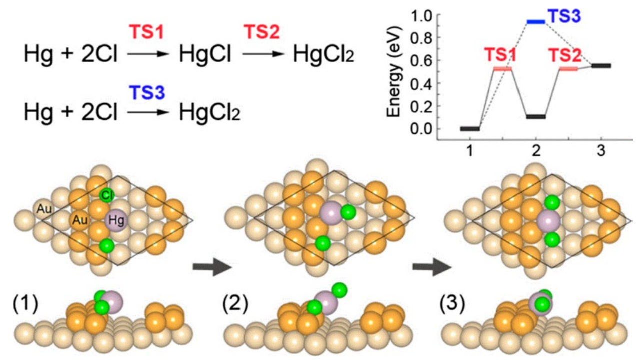

Fig. 2 Reaction pathways of mercury oxidation on the surface of Au. The Hg^0 oxidation on the surface of Au is a step-by-step Hg^0 oxidation (Hg → HgCl → $HgCl_2$) rather than a direct oxidation of Hg^0 to $HgCl_2$ (reproduced with permission from Lim and Wilcox 2013)

adsorption and removal efficiency, possibly due to the reaction of H_2S with the PdO to form PdS. Li et al. (2014a) suggested two Hg^0 removal pathways over the Pd/AC catalyst in N_2-Hg-H_2S atmosphere, reaction of Hg^0 with elemental palladium (Pd^0) to produce Hg-Pd amalgam or the reaction of some oxygen-containing functional groups on the surface of activated carbon (AC) with Pd^0 to form PdO. However, as shown in Fig. 1, the possible product in the N_2-Hg-H_2S atmosphere, PdS, is difficult to reduce to Pd^0, suggesting that this product could be inhibitory to the mercury removal process.

Gold-based catalysts are also considered as promising alternatives for Hg^0 removal because gold has the ability to adsorb and react with Hg^0 on its surface to form amalgam (Presto and Granite 2009; Zhao et al. 2006). Song and Lee (2016) synthesized a gold (Au)-based catalyst via an impregnation method and found the catalyst to achieve a 97% elemental mercury oxidation. Gómez-Giménez et al. (2015) studied the effect of SO_2 and O_2 on mercury removal and showed that these flue gas components promoted mercury removal in the presence of gold nanoparticles, attributable to the catalytic activity of Au. Ballestero et al. (2013) examined the regenerability of the Au-based catalyst through several cycles of Hg^0 capture regeneration and found that when the regeneration temperature was 220 °C, the Au-based catalyst maintained a high mercury removal efficiency in several regeneration cycles. In the process of elemental mercury oxidation, some reactants such as chlorine atoms have been shown to play an important role since gold could dissociate the adsorbed Cl_2 molecule into Cl atoms, which subsequently could react with Hg^0 to form $HgCl_2$, enhancing Hg^0 removal (Dranga and Koeser 2015). Lim and Wilcox (2013) examined the Hg^0 oxidation via a Langmuri–Hinshelwood (L–H) mechanism and suggested that the adsorbed Cl_2 (or HCl) could react with Hg^0 to produce HgCl and $HgCl_2$, as shown in Fig. 2, illustrating that the Hg^0 oxidation on the surface of Au is a step-by-step Hg^0 oxidation (Hg → HgCl → $HgCl_2$) rather than a direct oxidation of Hg^0 to $HgCl_2$.

Other noble metals such as Ag, Ru, and Ir also have been reported to be effective catalysts for mercury removal from flue gases (Karatza et al. 2011; Yan et al. 2011). Zhao et al. (2015d) prepared a Ag-based catalyst by an impregnation method and demonstrated its excellent performance for mercury removal in a simulated flue gas. Rungnim et al. (2015) synthesized Ag/TiO_2 catalyst samples by loading 5% Ag on TiO_2 powder and investigated possible synergistic effects between Ag and TiO_2 toward Hg^0 removal using periodic density functional theory (DFT) calculations. They showed an improved Hg^0 removal, suggesting the synergy resulted from the promotion of electron transfer from adsorbed elemental mercury to Ag/TiO_2 catalyst, with the concomitant effect of greatly enhancing the mercury removal.

It has been reported that RuO_2 is an excellent mercury oxidation catalyst and that halogen gases play an important role in the mercury oxidation process (Chen et al. 2014; Liu et al. 2016b, 2017). Liu et al. (2016b, 2017) studied the effect of halogen gas on mercury removal using RuO_2/TiO_2 catalyst in the presence of HCl or HBr, and the results showed 85 and 90% mercury removal in the presence of 10 ppm HCl and 1 ppm HBr, respectively, and that $HgCl_2$ and $HgBr_2$ were the main respective oxidation products. Liu et al. (2017) also found that the RuO_2/TiO_2 catalyst exhibited a good resistance to SO_2 poisoning under bituminous coal flue gas (SO_2 > 2000 ppm in flue gas). It was suggested that the oxidation reaction mechanism of elemental mercury follows the Deacon process as shown in Fig. 3. Chen et al. (2016) prepared the IrO_2-based catalyst via a sol–gel method and also found that the novel IrO_2-modified catalyst displayed a higher catalytic activity for mercury oxidation in a flue gas system, and the mechanism also followed the Deacon reacting scheme illustrated in Fig. 3.

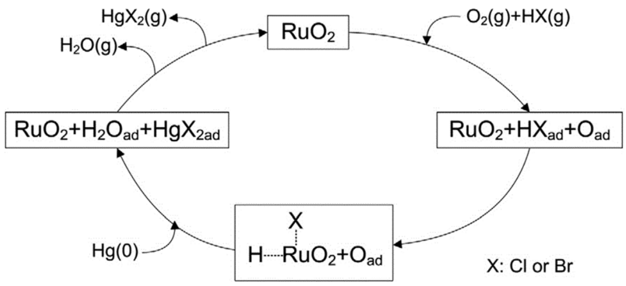

Fig. 3 Schematic diagram of Hg^0 oxidation reaction over RuO_2 catalyst in the presence of HCl or HBr. In the presence of HCl or HBr gas, the RuO_2 catalysts follow the Deacon process (reproduced with permission from Liu et al. 2016b)

Non-noble metal-based catalysts

Transition metal oxides-based catalysts

Transition metal oxides, including mainly Fe_2O_3, CuO, MnO_2, and CeO_2, commonly supported on carriers such as alumina, silica, titania, have been tested as potential elemental mercury oxidation catalysts. The advantages of these oxides compared with the noble metal catalysts, include the lower cost, widely available sources, and the relatively high catalytic oxidation activity. These supporters not only could increase the dispersion degree of metal oxides, but in some cases, also participate in the mercury removal process. Typical modification conditions and the resulting Hg^0 removal capacities are summarized in Table 2.

Copper-based catalysts are considered as promising mercury removal catalysts due to their abilities to store/release oxygen via the redox reaction between Cu^{2+} and Cu^+ (Tsai et al. 2013; Li et al. 2013c; Du et al. 2015). Liu et al. (2015b) synthesized Cu/Al_2O_3 catalyst via a wetness incipient method and reported that with optimal loading of 10 wt% Cu, more than 95% Hg^0 oxidation efficiency was attained during the first 20 h at 140 °C. It was also observed that the loading value of $CuCl_2$ has a significant effect on the activity of the catalyst. At low $CuCl_2$ loadings, it was speculated that $CuCl_2$ could react with Al_2O_3 to form copper aluminate ($CuAlO_2$) which was inactive for mercury oxidation, while high loadings of $CuCl_2$ would be expected to be present in a highly dispersed amorphous state on the surface of the $CuAlO_2$, which contributed to mercury removal. It was also observed that high loading of Cu into the Al_2O_3 support exhibited excellent SO_2 poisoning resistance under 10 ppm HCl (Yamaguchi et al. 2008). Zhou et al. (2014) tested the effect of HCl on Hg^0 removal using $CuCl_2/TiO_2$ catalyst, and they also found that the Cl atoms in HCl had a positive effect on Hg^0 removal. Xu et al. (2014a) suggested that CuO had showed a good performance for Hg^0 removal in the presence of low level HCl, and with a CuO/TiO_2 catalyst prepared by a wetness impregnation method, they reported Hg^0 removal efficiency of nearly 100% obtained with HCl concentration of 5 ppm. The positive effect of HCl was attributed mainly to the production of active atomic chlorine species.

Manganese-based catalysts are attractive potential alternatives for Hg^0 capture from flue gas due to their low cost and expected excellent oxidation performance, stemming from their inherent multiple oxidation states (Xu et al. 2015a; Li et al. 2010). Yu et al. (2015) investigated the performance of Hg^0 removal using M/Al catalysts (M = Mg^{2+}, Zn^{2+}, Cu^{2+}, and Mn^{2+}), and they found that compared with Mg/Al, Zn/Al, and Cu/Al catalysts, Mn/Al catalysts exhibited the highest Hg^0 removal performance at 300 °C. They concluded that Mn^{4+} species, which was the main active sites, played a very important role in the removal process of Hg^0. Xu et al. (2015b) reported that the improved removal of Hg^0 from flue gas, achieved with heterogeneous reaction between Hg^0 and Mn^{4+}, resulted from the transition of high valence (Mn^{4+}) to low valence Mn (Mn^{3+} and Mn^{2+}). Xie et al. (2013) also obtained similar results in the investigation of Hg^0 removal using Mn-based catalysts. Zhang et al. (2015a) examined the influence of calcination temperature in the 200–800 °C range on Hg^0 capture using MnO_x/TiO_2 sorbents. It was observed that the calcination temperature had an important effect on the activity and structure of the MnO_x/TiO_2 catalysts. The catalyst exhibited excellent performance for Hg^0 removal at high temperature of 400 °C; however, BET surface area, pore volume, and the content of Mn^{4+} of the catalyst decreased at calcination temperatures greater than 400 °C. Scala and Cimino (2015) studied the effect of flue gas composition on Hg^0 capture using manganese-based catalysts, and their results showed that both CO and CO_2 reduced the Hg^0 capture performance, while NO had no detectable effect, and 50 ppm HCl significantly improved the Hg^0 removal. Zhang et al. (2014a, 2015c) proposed that the Hg^0 oxidation by HCl over manganese-based catalyst followed the Hg → HgCl → $HgCl_2$ pathway, rather than the direct production of $HgCl_2$.

Scala and Cimino (2015) and Xie et al. (2012) examined the effect of SO_2 on Hg^0 removal both MnO_x-based and Mn-TiO_2 catalysts, and the results showed that SO_2 had a negative effects on the performance of both catalysts, mainly due to the competitive adsorption between Hg^0 and SO_2. Zhang et al. (2017b) also reported that the presence of SO_2 weakened the Hg^0 removal capacity of the MnO_x-based catalyst. To further enhance the Hg^0 removal effectiveness of Mn-based catalysts in the presence of SO_2, some metal elementals (Cu, Fe, Ce, Mo) have been utilized to modifying agents. Wang et al. (2013) prepared $CuO–MnO_2\_Fe_2O_3/Al_2O_3$ catalyst by an improved impregnation method and studied the effect of SO_2 concentration on Hg^0 removal, reporting that SO_2 has little effect on Hg^0 removal due to the larger affinity between Cu and sulfur. Zhao et al. (2016a) examined

Table 2 Reaction conditions and Hg^0 removal performance of non-noble metal-based catalysts

Raw sorbents	Name of modified sorbents	Simulated flue gas	CT* (°C)	RT** (°C)	MRE*** (%)	AC**** (μg/g)	References
Al_2O_3	$CuCl_2/\alpha-Al_2O_3$	$N_2/O_2/SO_2/HCl/Hg^0$	–	140	> 90	–	Li et al. (2013c)
Al_2O_3	$CuO_x-Al_2O_3$	$N_2/NO/SO_2/HCl/H_2O/CO_2/O_2/Hg^0$	500	140	> 65	–	Du et al. (2015)
$\gamma-Al_2O_3$	$CuCl_2/\gamma-Al_2O_3$	N_2/Hg^0	–	140	> 95	–	Liu et al. (2015b)
TiO_2	CuO/TiO_2	$N_2/HCl/O_2/Hg^0$	400	150	> 90	–	Zhou et al. (2014)
TiO_2	CuO/TiO_2	$O_2/HCl/N_2/Hg^0$	500	300	98	–	Xu et al. (2014a)
–	$Mn-Al-CO_3$	–	200	300	90	294.88	Yu et al. (2015)
–	$\alpha-MnO_2$	$N_2/O_2/Hg^0$	–	150	92	–	Xu et al. (2015b)
	$\beta-MnO_2$				> 10		
	$\gamma-MnO_2$				> 70		
–	$Zr_{0.5}Mn_{0.5}O_y$	$N_2/O_2/Hg^0$	500	100	–	5 mg/g	Xie et al. (2013)
TiO_2	Mn/Ti-200	$N_2/O_2/CO_2/Hg^0$	200	150	–	587	Zhang et al. (2015a)
	Mn/Ti-400		400			866	
$\gamma-Al_2O_3$	$MnO_x/\gamma-Al_2O_3$	$N_2/CO/CO_2/NO/O_2/SO_2/Hg^0$	550	50–250	–	–	Scala and Cimino (2015)
TiO_2	$Mn-TiO_2$	Air/Hg^0	400	300	95	1.6 mg/g	Xie et al. (2012)
TiO_2	Mn0.6Ti	$O_2/CO_2/HCl/H_2O/SO_2/NO/NH_3/Hg^0$	500	200	> 80	–	Zhang et al. (2017b)
$\gamma-Al_2O_3$	CMFA	$O_2/CO_2/HCl/NO/SO_2/H_2O/Hg^0/N_2$	500	300	> 70	–	Wang et al. (2013)
MoO_3/CNT	Mn-Mo/CNT	$O_2/N_2/SO_2/Hg^0$	400	150	> 90	–	Zhao et al. (2016a)
$\gamma-Al_2O_3$	MnCe	$O_2/CO_2/HCl/NO/SO_2/H_2O/Hg^0/N_2$	550	250	> 80	–	Wang et al. (2014)
TiO_2	CeTi	$O_2/H_2O/CO_2/HCl/NO/SO_2/N_2/Hg^0$	500	250	> 90	–	Li et al. (2011b)
TiO_2	CeTi	$HCl/NO/O_2/Hg^0/N_2$	500	200	96.1	–	Li et al. (2013a)
CS	MnO_2/CeO_2-MnO_2	$O_2/N_2/Hg^0$	450	150	89	–	Ma et al. (2017)
TiO_2	CeTi	$H_2/CO/H_2S/HCl/NH_3/N_2/Hg^0$	500	120	> 80	–	Zhou et al. (2013)
TiO_2	CeTi	$H_2/CO/H_2S/HCl/N_2/Hg^0$	500	150	> 80	–	Hou et al. (2014b)
TiO_2	VCeTi	$O_2/N_2/Hg^0$	500	250	81.55	–	Zhang et al. (2015e)
TiO_2	CuCeTi	$NO/NH_3/O_2/N_2/Hg^0$	500	200	90	–	Li et al. (2017d)
$V_2O_5-WO_3/TiO_2$	VWTiCe	$NO/SO_2/O_2/CO_2/N_2/Hg^0$	500	250	88.93	–	Zhao et al. (2015c)
Selective catalytic reduction (SCR)	$WO_3-V_2O_5/TiO_2$	$N_2/O_2/HCl/Hg^0$	–	350	98.5	–	Gao et al. (2013a)
TiO_2	$V_2O_5-WO_3/TiO_2$	$CO_2/O_2/N_2/Hg^0$	500	250	> 80	–	Wang et al. (2015a)
TiO_2	TVM	$O_2/N_2/Hg^0$	400	350	> 80	–	Zhao et al. (2014)
TiO_2	TV_5M_5	$O_2/CO_2/CO/NO/NO_x/SO_2/H_2O/HCl/Hg^0$	400	370	> 90	–	Zhao et al. (2015a)
Selective catalytic reduction (SCR)	Fe_2O_3/SCR	$HCl/O_2/N_2/Hg^0$	400	350	> 90	–	Huang et al. (2016)
	Ce-Cu/SCR	$N_2/O_2/NO/NH_3/Hg^0$	500	250	> 80	–	Chi et al. (2017)
	MnO_x 5%/catalyst	$CO_2/H_2O/O_2/HCl/SO_2/NO/NH_3/N_2/Hg^0$	–	350	83.8	–	Chiu et al. (2015)

*Calcination temperature

**Reaction temperature

***Hg^0 removal efficiency

****Adsorption capacity

the effect of SO_2 on Hg^0 capture using Mo-doped Mn/CNT catalyst, showing that the presence of SO_2 improved Hg^0 removal, and attributing this to Mo promoting of the conversion of SO_2 to SO_3, with concomitant improvement in Hg^0 removal efficiency. Wang et al. (2014) also investigated the effect of SO_2 on Hg^0 removal using MnO_x–CeO_2/Al_2O_3 catalyst, and found that the addition of Ce effectively resisted the poisoning effect of SO_2 on the catalysts.

Cerium oxide and Ce-based catalysts have gained widespread attention due to the unique redox cycle between Ce^{4+} and Ce^{3+}, excellent oxygen storage capacity and high oxidation capacity, and resistant to SO_2 poisoning (Li et al. 2011b). In the process of redox reaction of Ce^{4+} and Ce^{3+}, bulk oxygen species and surface oxygen vacancies with high mobility are easily produced, which facilitate the effectiveness of Hg^0 removal. Li et al. (2013a) synthesized Ce-based catalysts using TiO_2 nanoparticles by an ultrasonic-assisted impregnation method, and reported that the addition of 1200 ppm SO_2 into a flue gas system enhanced the performance of Hg^0 capture. In addition, the results of Ma et al. (2017) showed that the addition of CeO_2 improved the water vapor resistance of the catalyst, and even with 5% water vapor in the flue gas, the high-level removal efficiency of Hg^0 was only slightly reduced. Considering the superior activity and the unique redox cycles of Ce^{4+}/Ce^{3+}couple, the incorporation of CeO_2 into other metal oxide catalysts is generally believed to improve their Hg^0 removal performances.

Zhou et al. (2013) and Hou et al. (2014b) investigated the Hg^0 removal over CeO_2–TiO_2 catalysts and reported that when HCl or H_2S was present alone in the flue gas, more than 97% of Hg^0 was captured, while the simultaneous presence of HCl and H_2S resulted a prohibitive effect on the effectiveness of Hg^0 capture. Zhou et al. (2013) also found that the presence of H_2 and CO have a negligible effect on the capture of Hg^0 at 150 °C. Zhang et al. (2015e) synthesized a series of Ce-based V_2O_5/TiO_2 catalysts by an ultrasound-assisted impregnation method and found that the V(1) Ce(10)Ti catalyst had the best Hg^0 oxidation performance. Li et al. (2017d) examined the synergistic effect of CeO_2 and CuO using CuTi, CeTi and CuCeTi catalysts prepared by a sol–gel method. They found that, unlike the CuTi and CeTi catalysts, the Hg^0 removal efficiency of CuCeTi catalyst at 200 °C was about 99.0%, the high value ascribed to the combined effect of the presence of both CeO_2 and CuO.

Selective catalytic reduction catalysts

Recently, selective catalytic reduction (SCR) systems have been applied in many coal-fired power plants for NO_x removal due to its higher economy of scale, efficiency, and selectivity. Typical selective catalytic reduction (SCR) catalysts usually apply TiO_2 and some catalytically active components (such as WO_3, V_2O_5 and/or MoO_3) as precursors and activators, respectively. The modification conditions and Hg^0 removal capacities of SCR type catalysts are summarized in Table 2. V_2O_5 is the major active ingredient of the selective catalytic reduction (SCR) catalyst, which can be employed not only to control the emission of NO_x but also to remove Hg^0 from flue gas. Zhao et al. (2015c) reported that the V_2O_5-rich SCR catalyst exhibited a superior Hg^0 removal performance in the range of 250–350 °C. For WO_3–V_2O_5/TiO_2 catalysts, the Hg^0 oxidation in the presence of both O_2 and HCl was found to follow the Eley–Rideal mechanism (Gao et al. 2013a). Wang et al. (2015a) also investigated the Hg^0 removal in CO_2-enriched flue gas using WO_3–V_2O_5/TiO_2 catalysts, and they found that high concentration of CO_2 (80 vol%) promoted the capture efficiency of Hg^0, but inhibited the removal of NO. MoO_3 is often introduced into the catalyst's formulation to improve its resistance to SO_2 poisoning. Zhao et al. (2014) found that the V_2O_5–MoO_3/TiO_2 catalyst was excellent Hg^0 oxidation, and the Hg^0 removal process could be explained by the Mars–Maessen mechanism. To further study the Hg^0 capture performance of this catalyst system in actual flue gas, Zhao et al. (2015a) performed a test in a coal-fired power plant, and reported higher than 90% Hg^0 removal efficiency.

Selective catalytic reduction (SCR) system is widely applied in coal-fired power plant to simultaneously control the emissions of NO_x and Hg^0. However, the conventional selective catalytic reduction catalysts are not effective enough for the removal of Hg^0 in the presence of low HCl concentrations and are often suppressed by the presence of SO_2 and NH_3 in the flue gas (Kamata et al. 2008). Therefore, some metal oxides are usually used to modify the selective catalytic reduction (SCR) catalysts. Huang et al. (2016) prepared the Fe_2O_3/SCR catalyst by an impregnation method and found that the introduction of Fe_2O_3 could significantly improve the Hg^0 removal ability of the SCR catalyst. The active temperature window of Fe_2O_3/SCR catalyst was found to range from 150 to 450 °C, which is wider than that of conventional SCR catalysts. They suggested that the Fe^{3+} could react with HCl to release active Cl species by the Mars–Maessen mechanism and then the generated active Cl species could participate in the Hg^0 removal by the L–H mechanism. The proposed plausible Hg^0 oxidation mechanism is shown in Fig. 4.

Chi et al. (2017) prepared a series of Ce-Cu-modified selective catalytic reduction (SCR) catalysts by ultrasonic-assisted impregnation method for simultaneous removal of Hg^0 and NO_x and found that a 7%Ce–1%Cu/SCR catalyst showed a superior performance at 200–400 °C. The catalyst also exhibited higher resistance to water vapor and SO_2. The Hg^0 removal performance of MnO_x-treated commercial SCR catalysts was also evaluated (Chiu et al. 2015), and the

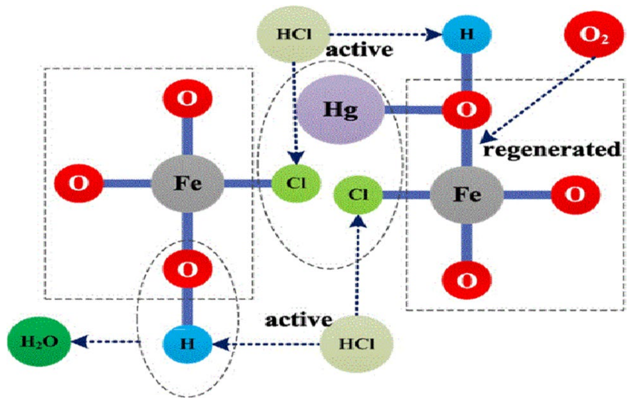

Fig. 4 Schematic of the possible Hg^0 oxidation mechanism in HCl-O_2 on over the Fe_2O_3/SCR catalyst. The active chlorine species generated by the reaction of Fe^{3+} and HCl can react with adsorbed Hg^0 to form $HgCl_2$. The gas-phase O_2 in flue gas regenerated the chemisorbed oxygen and lattice oxygen (reproduced with permission from Huang et al. 2016)

results showed that both 5 and 10% MnO_x-impregnated SCR catalysts had higher Hg^0 oxidation efficiency.

Activated carbon/cokes based sorbents

Activated carbon/cokes have been proven to be effective sorbents for Hg^0 removal, and sulfur, halogens, and metal oxides are the most common additives/modified reagents, which have been widely studied for the modifications of these sorbents to improve their removal efficiencies for Hg^0. The modification conditions and Hg^0 removal capacities of activated carbon-/coke-based sorbents are summarized in Table 3.

Sulfured carbon sorbents

Sano et al. (2017) performed a laboratory-scale test of Hg^0 removal over sulfur-impregnated activated carbon and raw activated carbon and reported that S (sulfur) impregnation resulted in 50 times higher Hg^0 removal than the performance of the raw activated carbon. Hsi and Chen (2012) studied the effects of acidic/oxidizing gases, O_2, HCl, SO_2, and NO which are commonly found in the flue gas, on Hg^0 removal using simulated flue gas over sulfur-impregnated activated carbon. They observed the flue gas components had strong positive effect on the catalyst's performance, with the largest Hg^0 removal capacity of 2310 μg/g obtained in the presence of O_2, HCl and NO.

Ie et al. (2013) synthesized a series of innovative composite powdered activated carbon (PACs) by an impregnation method using aqueous-phase sodium sulfide (Na_2S) and vapor-phase elemental sulfur (S^0) in different sequences and investigated their performances in the removal of Hg^0 or $HgCl_2$. They found that the Hg^0 and $HgCl_2$ removal capacities of powdered activated carbon (PACs) impregnated with aqueous Na_2S solution followed by gaseous sulfur (S^0), respectively, were 1.98 and 1.42 times higher than those of the samples impregnated in the opposite sequence. Yao et al. (2014) also studied the performance of activated carbon fibers functionalized with sulfur-containing groups and reported that sulfur impregnation decreased the pore volume and surface area of activated carbon fibers. However, compared with the raw activated carbon fiber samples, the Hg^0 removal capacity of sulfur-treated activated carbon fibers increased due to the incorporation of the sulfur groups.

Halogenated carbon sorbents

Zhou et al. (2015) prepared Br-based activated carbon by an impregnation method and evaluated the in-flight Hg^0 capture performance in an entrained flow reactor. They found that the Hg^0 removal efficiency of raw activated carbon was significantly enhanced by the NH_4Br impregnation. Yao et al. (2013) also prepared Br-based activated carbon fibers using KBr solution, and by KBr impregnation, bromine vapor, and electrochemical modification methods, respectively. For the brominated activated carbon fibers, the introduction of Br atoms promoted the Hg^0 oxidation process. They also found that the brominated activated carbon fibers modified by bromine vapor and electrochemical methods using KBr solution exhibited stable Hg^0 removal capacity (30–33% capture), which was retained up to 3 months. Rupp and Wilcox (2014) examined the effects of flue gas components (NO_x, SO_2) on Hg^0 removal using brominated activated carbon fibers and reported that while NO_x promoted the oxidation of Hg^0, SO_2 prevented the Hg^0 adsorption, and the interaction of NO_x and SO_2 with Br decreased sorbent's performance.

Tsai et al. (2017) investigated $CuCl_2$-impregnated activated carbon for Hg^0 removal using a fixed-bed reactor system. Results from the tests showed that the Cl-impregnated samples achieved better Hg^0 removal capacity than non-impregnated samples, with the Hg^0 removal capacity of the 8% $CuCl_2$-impregnated sample reported to be 631.1 μg/g. Li et al. (2017a) prepared NH_4Cl-modified activated carbons for Hg^0 removal by an impregnation method, and they found that Cl-doped activated carbons exhibited a good performance for Hg^0 capture. De et al. (2013) impregnated activated carbons using various halogens such as ammonium and potassium halides. They observed that the introduction of halide ions greatly enhanced the capacity of the activated carbons for Hg^0 removal. For the same loading values of halide (I, Cl and Br) ions, the Hg^0 capture performance of ammonium halide-modified activated carbon was higher than those of potassium halide-modified activated carbon. Also, the I-impregnated sample exhibited the highest Hg^0

Table 3 Reaction conditions and Hg^0 removal performance of carbon sorbents

Raw sorbents	Name of modified sorbents	Simulated flue gas	RT* (°C)	MRE** (%)	AC*** (mg/g)	References
Activated carbon (AC)	AC-S	Air/Hg^0	140	–	21	Sano et al. (2017)
AC	AC-400S	$CO_2/H_2O/O_2/HCl/N_2/NO/Hg^0$	150	–	2.31	Hsi and Chen (2012)
Powdered activated carbon (PAC)	Na_2S+S^0-PAC	N_2/Hg^0	150	–	33.789	Ie et al. (2013)
ACF	NaSH-ACF	Air/Hg^0	–	–	> 14	Yao et al. (2014)
AC	NH_4Br-AC	$O_2/CO_2/CO/NO/SO_2/Hg^0$	–	90.5	–	Zhou et al. (2015)
Activated carbon fiber (ACF)	Br(v)-ACF	Air/Hg^0	–	–	64	Yao et al. (2013)
	KBr-ACF				100	
	eBr-ACF				50	
	Br-ACF	$CO_2/H_2O/N_2/O_2/Hg^0$	140	97	–	Rupp and Wilcox (2014)
AC	HCAC	N_2/Hg^0	150	–	0.631	Tsai et al. (2017)
AC	ACNCl5	$O_2/SO_2/HCl/NO/N_2/Hg^0$	180	71.9	–	Li et al. (2017a)
AC	KCl-AC	N_2/Hg^0	135	26	–	De et al. (2013)
	KBr-AC			85	–	
	KI-AC			100	–	
	NH_4Br-AC			> 50	–	
	NH_4I-AC			100	–	
AC	KI-AC	N_2/Hg^0	120	> 90	–	Tong et al. (2017)
AC	CuO/AC-Hz	$N_2/O_2/Hg^0$	120	> 72	–	Zhao et al. (2016b)
Semi-coke (SC)	Ce/SC	Air/N_2/Hg^0	260	95	–	Zhang et al. (2017a)
SC	Ce/SC	Air/N_2/Hg^0	260	> 95	–	Zhao et al. (2017a)
AC	CeO_2–Mn/AC	N_2/Hg^0	120	90	–	Wu et al. (2017)
AC	MnCe/AC	$N_2/O_2/Hg^0$	190	> 90	–	Xie et al. (2015)
SC	Mn/Ce-SC	$N_2/O_2/Hg^0$	260	> 90	–	Zhang et al. (2016d)
AC	CoCe/AC	$O_2/CO_2/NO/SO_2/N_2/Hg^0$	170	> 80	–	Wu et al. (2015b)
AC	Fe_2O_3–CeO_2/AC	$O_2/CO_2/NO/SO_2/N_2/Hg^0$	110	90	–	Wang et al. (2016c)
AC	AC-A30	N_2/Hg^0	30	> 70	–	Zhang et al. (2015b)
AC	AC-O20	N_2/Hg^0	25	> 50	37.05 μg/g	Zhang et al. (2016e)
AC	AC-C15T60	N_2/Hg^0	30	> 96	–	Zhang et al. (2016c)

*Reaction temperature

**Hg^0 removal efficiency

***Adsorption capacity

removal capacity compared to Cl- and Br-impregnated samples.

Tong et al. (2017) synthesized the I-impregnated activated carbons using an impregnation method and investigated the Hg^0 capture, and the adsorption mechanism and the effects of simulated flue gas components. They found that the formation of I_2 molecules on the surface of I-impregnated activated carbons significantly promoted Hg^0 removal and proposed the plausible adsorption mechanism shown in Fig. 5. They also observed that low concentrations of SO_2 had a promotional effect on Hg^0 oxidation, but high concentrations of SO_2 had a negative impact on Hg^0 capture. They also found that the Hg^0 removal efficiency significantly increased with increasing NO concentration from 0 to 100 ppm, while high NO concentration of 300 ppm showed antagonistic effects.

Metal oxides-modified carbon sorbents

Zhao et al. (2016b) studied the use of activated coke impregnated with CuO (a CuO/AC-H sample), focusing on the effects of the copper loading, reaction temperature, calcination temperature, and flue gas components (NO, O_2) on Hg^0 capture, and found the optimal reaction temperature, copper loading value, and calcinations temperature to be 160 °C, 8%, and 300 °C, respectively, and that NO and O_2 showed positive effects on Hg^0 capture. CeO_2 has been widely investigated as one of the catalysts for selective

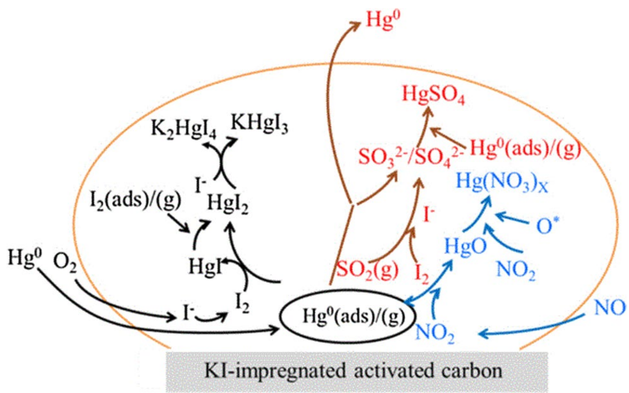

Fig. 5 Possible adsorption mechanism for Hg^0 under simulated flue gas. The formation of I_2 molecules, SO_3^{2-}/SO_4^{2-} active species and NO_2 active species on the surface of I-impregnated activated carbons significantly promoted Hg^0 removal (reproduced with permission from Tong et al. 2017)

catalytic reduction (SCR) of NO_x, and Hg^0 removal due to its large oxygen storage capacity and unique redox couple Ce^{3+}/Ce^{4+}, and excellent ability to shift between CeO_2 and Ce_2O_3 under oxidizing and reducing conditions, respectively (Zhang et al. 2017a; Zhao et al. 2017a). Zhang et al. (2017a) prepared CeO_2-supported semi-coke (SC) sorbents by an impregnation method and observed much better Hg^0 removal capacity than that of unmodified semi-coke (SC) but high concentration of H_2O vapor showed inhibitory effects. It was demonstrated that the Ce–OH groups formed by the reaction of CeO_2 and H_2O vapor consumed lattice oxygen on the surface of samples, with the concomitant effect of decreasing the Hg^0 removal efficiency. Zhao et al. (2017a) also obtained similar results in studying the effect of water vapor on Hg^0 removal performance over CeO_2-supported semi-coke (SC) sorbents. Wu et al. (2017), Xie et al. (2015), and Zhang et al. (2016d) prepared Ce–Mn-co-modified activated carbons (AC), Mn–Ce-mixed oxides-modified activated coke (MnCe/AC), and Mn/Ce-modified semi-coke (Mn/Ce-SC) by an impregnation method, respectively, and found the modified sorbents to exhibit excellent Hg^0 capture capability. Wu et al. (2015b) investigated the performance of CoCe/AC sorbents prepared by an impregnation method for Hg^0 capture from flue gas at 110–230 °C and reported superior performance compared to Ce/AC, Co/AC, and virgin AC, with a 92.5% Hg^0 removal achieved at 170 °C. Based on the results obtained from XPS and TGA analyses, the valence transitions of Co^{3+}/Co^{2+} and Ce^{4+}/Ce^{3+} produced lattice oxygen, promoting Hg^0 oxidation and removal. Wang et al. (2016c) also reported that activated coke (AC) impregnated with CeO_2 and Fe_2O_3 (denoted Fe_2O_3-CeO_2/AC), significantly improved Hg^0 removal capacity.

Plasma-treated carbon sorbents

In recent years, plasma modification has received widespread attention in research for functionalizing catalyst and sorbents. For example, non-thermal plasma produces energetic electrons, ions, and active radicals, which could improve the pore structure of sorbents and increase the active functional groups on the surface of the sorbents. Some investigators (Zhang et al. 2015b, 2016c, e) have shown that plasma modification could form multiple functional groups on the surface of sorbents, ameliorating the Hg^0 removal process.

Zhang et al. (2015b) used a non-thermal plasma technology to modify activated carbon (AC) in air environment and found the modified sample to have a higher Hg^0 removal efficiency than the corresponding raw sample. The results of XPS showed that the modification by non-thermal plasma increased the content of ester groups (C(O)–O–C) and carbonyl groups (C=O) on the activated carbon, which played a key role in Hg^0 removal. Zhang et al. (2016e) also obtained the similar results in studying the effect of oxygen non-thermal plasma modification, reporting that the modified activated carbon (AC) exhibited a high removal performance for Hg^0 from flue gas. Zhang et al. (2016c) modified activated carbon (AC) by Cl_2 non-thermal plasma method and found the sample to greatly enhance Hg^0 removal by increasing the chlorinated (Cl) active sites on the surface of the activated carbon (AC). The corresponding XPS analysis indicated that a large number of C–Cl groups resulted from the treatment, which could have oxidized the Hg^0 to $HgCl_2$, as illustrated in Fig. 6.

Biomass char-based sorbents

The results of some studies (Hua et al. 2010; Lee et al. 2006; Diamantopoulou et al. 2010) have indicated that injection of activated carbon into the flue gas system is a promising method for Hg^0 removal from flue gas. However, large activated carbon (AC)/Hg^0 ratio and high operation costs have limited large-scale applications (Hsi et al. 2002; Scala et al. 2011). Biomass char is the by-product of biomass pyrolysis under oxygen-free conditions. With the low costs and the simplicity of preparation, it could be considered as an attractive alternative to activated carbon (AC) (Liu et al. 2011). Therefore, pyrolysis chars, which are made from cheap and renewable resources, have been extensively studied recently in the field of Hg^0 removal (Hsi et al. 2011; Klasson et al. 2010; Fuente-Cuesta et al. 2012). However, they require physical techniques to modify pore structure such as specific surface area, pore volume, and pore size and/or chemical modification to increase active functional groups on the surface. For example, the use of active ingredients such as halogens, metal oxides, and acid to modify biochars has

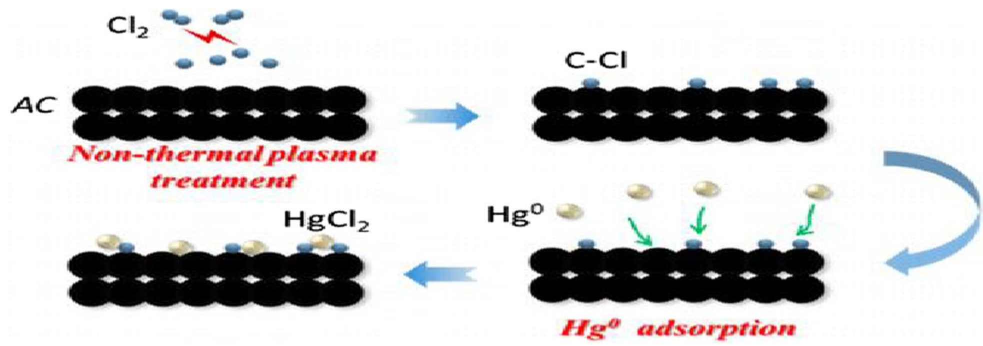

Fig. 6 Mechanism of modified activated carbon (AC) for Hg^0 removal. A large number of C–Cl groups generated by the Cl_2 non-thermal plasma treatment can oxidize Hg^0 to $HgCl_2$, thus promoted the removal of Hg^0 (reproduced with permission from Zhang et al. 2016c)

been reported to improve their performance for Hg^0 removal. The modification conditions and Hg^0 removal capacities of modified biochars are summarized in Table 4.

The results of a number of studies (Johari et al. 2016a, b; Klasson et al. 2014) have suggested that the pyrolysis conditions could also substantially influence the yield and physicochemical properties of chars. Johari et al. (2016b) prepared a series of coconut pith (CP) chars at different pyrolysis temperatures and found the Hg^0 removal capacity to increase with increasing pyrolysis temperature, with the highest removal capacity of 6067.49 µg/g obtained at 900 °C. Klasson et al. (2014) prepared four different biochars (almond shells, cottonseed hulls, lignin, and chicken manure) at different pyrolysis temperature and reported that chicken manure exhibited the best Hg^0 removal performance of 95% from flue gas at 650 and 800 °C.

Halogens-modified biochar

Many studies have reported that chemical modifications of sorbents using halogens could significantly enhance the Hg^0 removal from flue gas (Li et al. 2015a, b; Shen et al. 2015a). The halogens (chloride, bromide, and iodide) have been demonstrated as effective reagents for modification of sorbents to improve their performance in Hg^0 removal from flue gas. Li et al. (2015a) and Shen et al. (2015a) prepared low-cost sorbents using municipal solid waste and medicinal residues by a chloride impregnation method and reported that NH_4Cl-modified sorbents showed improved performance for Hg^0 removal. Li et al. (2015b) investigated the effects of flue gas composition on Hg^0 removal using NH_4Cl-impregnated medicine residue biochars and reported that the presence of O_2 and NO increased Hg^0 removal, but water vapor suppressed the removal process. A dual effect of SO_2 concentration was observed on Hg^0 capture, that is, low SO_2 concentration enhanced Hg^0 removal while high SO_2 concentration was antagonistic.

Li et al. (2015c) carried out a comparative study of NH_4Cl modified biochars from three solid wastes (medicinal residues, municipal solid wastes, and cotton straw), showing that the chemically modified biochars, especially the modified cotton straw char exhibited higher Hg^0 removal capacity than modified activated carbon (AC). In addition, the biochar derived from waste tire also demonstrated an excellent Hg^0 removal performance, resulting from generated mercury sulfide chemisorption sites on the surface of the biochar (Li et al. 2017b). Shen et al. (2017) studied NH_4Cl-modified biochar sorbents derived from waste tea and found that the generated C–Cl and C=O groups on the surface of the biochar promoted the oxidation of Hg^0, resulting in an excellent Hg^0 removal. Xu et al. (2016c) synthesized a novel Cl-Char composite by the co-pyrolysis of biomass (wood and paper) and polyvinyl chloride (PVC) and reported a 90% Hg^0 removal capacity at 140 °C, which was more than 2.5–5 times than that of a raw char. In addition, biochars modified by metal chlorides have been evaluated for their Hg^0 capture performances from flue gas (Shu et al. 2013; Tan et al. 2015). Shu et al. (2013) studied mulberry twig chars (MT) modified by $ZnCl_2$, H_2O_2, and NaCl, respectively, and reported that the $ZnCl_2$-impregnated char was better than the other chemically treated samples for Hg^0 removal. Tan et al. (2015) also compared Hg^0 capture performance of bamboo charcoal (BC) impregnated by $ZnCl_2$ and $FeCl_3$ and found that the impregnated BCs was better than raw bamboo charcoal (BC), with the $FeCl_3$-impregnated BCs showing the highest Hg^0 removal efficiency (> 99.9%) at 140 °C.

It is well known that chemical modification of sorbents with bromine plays a key role in the adsorption and oxidation of Hg^0 (Yang et al. 2018a, b; Tang et al. 2017; Zhu et al. 2016). Yang et al. (2018a, b) reported that sargassum chars' effectiveness for Hg^0 removal was greatly improved after NH_4Br and NH_4Cl impregnation, with the NH_4Br-modified samples showing improved performance attributable to the generation of C–Br and C=O groups on the surface of the sorbents. Tang et al. (2017) developed a low-cost sorbent based on rice husk char (RHC) using HBr impregnation method and demonstrated that the modified rice husk char (RHC-HBr) had higher Hg^0 removal capacity (57.84 µg/g) than those of activated carbon (AC). Zhu et al. (2016) evaluated the performances of chemically treated samples of rice husk char (RHC) and commercial coal-based activated carbon (CAC) and found that modification of

Table 4 Reaction conditions and Hg^0 removal performance of biochars

Raw sorbents	Name of modified sorbents	Simulated flue gas	PT* (°C)	RT** (°C)	MRE*** (%)	AC**** (μg/g)	References
Biomass chars	PW1	$O_2/SO_2/NO_2/HCl/Hg^0/N_2$	–	150	–	172	Fuente-Cuesta et al. (2012)
Coconut pith chars	CP700	N_2/Hg^0	700	–	–	2395.98	Johari et al. (2016a, b)
	CP900		900			6067.49	
Chicken manure	/	$HCl/NO_x/SO_2/O_2/CO_2/Hg^0/N_2$	800	150	–	250	Klasson et al. (2014)
Municipal solid waste	C6WN5	$N_2/O_2/Hg^0$	600	80	–	157.7	Li et al. (2015a)
Medicine residue	M6WN5	$N_2/O_2/Hg^0$	600	120	–	869.6	Shen et al. (2015a) and Li et al. (2015b)
Municipal solid wastes	W6WN5	$N_2/O_2/Hg^0$	600	120	–	160	Li et al. (2015c, 2017b)
Cotton straw char	C6WN5					11400	
Medicinal residues	M6WN5					840	
Waste tire	T6WN5			–	83.2	–	
Waste tea	HCU-5	$N_2/O_2/Hg^0$	500	120	> 90	–	Shen et al. (2017)
Paper/Wood	Paper/PVC	N_2/Hg^0	700	140	90	–	Xu et al. (2016c)
	Wood/PVC						
Mulberry twig chars	MT873-A-Z5	$N_2/NO/SO_2/Hg^0$	600	90	–	29.55	Shu et al. (2013)
Bamboo charcoal	B1/B2	$O_2/SO_2/NO/CO_2/Hg^0/N_2$	–	140	88/92	–	Tan et al. (2015)
	B3/B4				99.9		
Sargassum chars	S8Br5	$N_2/SO_2/NO/O_2/H_2O/Hg^0$	800	160	93.96	952.4	Yang et al. (2018a, b)
	S8Cl5				91.67	625.0	
Rice husk char	RHC-HBr	$O_2/CO_2/SO_2/NO/N_2/Hg^0$	600	150	–	57.84	Tang et al. (2017)
Rice husk char	RBr	$N_2/SO_2/NO/Hg^0$	600	150	> 70	> 30	Zhu et al. (2016)
	RCl				> 60	> 25	
Bamboo charcoal	BC-I	$O_2/SO_2/NO/CO_2/Hg^0/N_2$	–	140	99.9	–	(Tan et al. 2012b)
				180	> 90	–	
Sargassum chars	S8KI3	$N_2/SO_2/NO/O_2/H_2O/Hg^0$	800	160	94.1	–	Liu et al. (2018)
Enteromorpha chars	E8KI3				95.7		
Cotton straw char	C6WNCl1	N_2/Hg^0	600	120	–	1239.2	Li et al. (2016a, 2017c)
	C6WNBr1					2781.9	
	C6WNI1					7752.0	
Sawdust	$Fe_{1.5}MBC_{600}$	$N_2/O_2/Hg^0$	600	120	> 90	1279.6	Yang et al. (2016a)
Wheat straw char	WS8Fe0.1	$N_2/H_2O/O_2/Hg^0$	600	50	> 80	–	Zhou et al. (2017)
Wheat straw char	MnCe0.12(2/1)/WSU250	$N_2/H_2O/O_2/NO/SO_2/Hg^0$	600	150	83.6	–	Yang et al. (2017b)
Rice straw char	CuCe0.18(1/5)/RSU(260)	$N_2/H_2O/O_2/NO/SO_2/Hg^0$	600	150	79.93	–	Xu et al. (2018)
Peanut shells	6Mn-6Zr/PSC-I3	$N_2/O_2/Hg^0$	600	150	> 90	5587.0	Zeng et al. (2017)
Bamboo char	BC2	$N_2/O_2/Hg^0$	–	160	> 70	294.1	Xu et al. (2016a)
Waste tire	T6 N	$N_2/O_2/Hg^0$	600	120	–	–	Li et al. (2015d)
	T6S						
Leather industry waste	BCT0.33	$N_2/O_2/Hg^0$	750	150	–	2007	Lopez-Anton et al. (2015)
Corn stalk char	BC-50-9	N_2/Hg^0	700	140	–	269.4	Niu et al. (2017)
Tobacco straw	T6Cl	Air/Hg^0	600	150	84.2	583.0	Wang et al. (2018)
Rice straw	R6Cl				83.9	445.1	
Millet straw	M6Cl				81.1	444.3	
Wheat straw	W6Cl				52.7	217.6	
Corn straw	C6Cl				42.7	150.8	
Black bean straw	B6Cl				5.3	12.6	

*Pyrolysis temperature

**Reaction temperature

***Hg^0 removal efficiency

****Adsorption capacity

NH_4Cl and NH_4Br significantly increased the Hg^0 removal efficiency of rice husk char compared to CAC and that the NH_4Br-modified rice husk char exhibited the highest Hg^0 removal performance.

Tan et al. (2012b) reported that KI modification of bamboo charcoal (BC) by an impregnation method, while it resulted in the decrease in the total volume and BET surface area, the modified BC (BC-I) exhibited superior capacity for Hg^0. The results of XPS analysis of the used samples appear to support the generations of C-I_x compounds and I_2 and subsequent reactions with Hg^0 to form iodated mercuric compounds, thus contributing to a higher Hg^0 removal efficiency. Liu et al. (2018) also obtained similar results in their study of the removal of Hg^0 using the KI-modified sargassum and enteromorpha chars. Li et al. (2016a, 2017c) synthesized cotton straw char sorbents using three different ammonium halides to capture Hg^0 from flue gas and found that the Hg^0 removal efficiency was in the order of $NH_4I > NH_4Br > NH_4Cl$. It was also noted that high reaction temperature improved the Hg^0 removal performance of the NH_4I-modified sorbents.

Metal oxides-modified biochar

In recent years, metal oxides have been widely studied as effective sorbent modifiers for Hg^0 capture due to their low costs and high activities. Among metal oxides used to modify biochars-based sorbents for Hg^0 capture are FeO_x, CeO_x, CuO_x, MnO_x, and ZrO_2 (Yang et al. 2016a, 2017b; Zhou et al. 2017; Xu et al. 2018; Zeng et al. 2017). Yang et al. (2016a) prepared a novel magnetic sorbents (MBC) based on sawdust char by one-step pyrolysis of $FeCl_3$-laden method and showed that the modified sample has improved Hg^0 removal capacity compared with those of raw biochar. XPS analysis indicated that the generated Fe_3O_4 and C=O groups were the major active oxidation/adsorption sites for Hg^0 removal. The plausible mechanism of Hg^0 removal proposed is depicted in Fig. 7.

Zhou et al. (2017) studied Hg^0 removal by wheat straw char impregnated with K_2FeO_4 reagent, and the results appeared to show that K_2FeO_4 impregnation effectively improved pore structure of the wheat straw char, leading to enhancement in Hg^0 removal. Yang et al. (2017b) further studied the Hg^0 removal performance of wheat straw char modified by Mn–Ce-mixed oxides and found that the Mn/Ce redox cycle played an important role in Hg^0 removal. Xu et al. (2018) modified rice straw char (RS) by impregnation with Cu-Ce-mixed oxides to remove Hg^0 from flue gas and reported significant enhancement up to 95.26% efficiency. Zeng et al. (2017) prepared metal oxides (MnO_x and ZrO_2) and halide ions (I^-) modified peanut shells char (6Mn-6Zr/PSC-I3) and demonstrated that the sample exhibited superior Hg^0 removal capacity (15028.4 μg/g). Based on XPS analysis, two reaction stages could be detected in the Hg^0 removal process. As shown in Fig. 8, at the initial reaction stage, Hg^0 was first removed by the chemical adsorption sites of C-I groups. The Hg^0 oxidation caused by the hydroxyl (OH) oxygen and lattice oxygen played a key role at the final reaction stage.

Other modification

In addition to the modification of biochar sorbents with halogens and metal oxides, other chemical modification methods involving the use of acid and alkali to increase surface activity, and physical modification such as plasma mainly to

Fig. 7 Mechanism of magnetic sorbents for Hg^0 removal. The generated Fe_3O_4 and C=O groups on the surface of novel magnetic sorbents (MBC) by one-step pyrolysis of $FeCl_3$-laden method significantly promoted Hg^0 removal (reproduced with permission from Yang et al. 2016a)

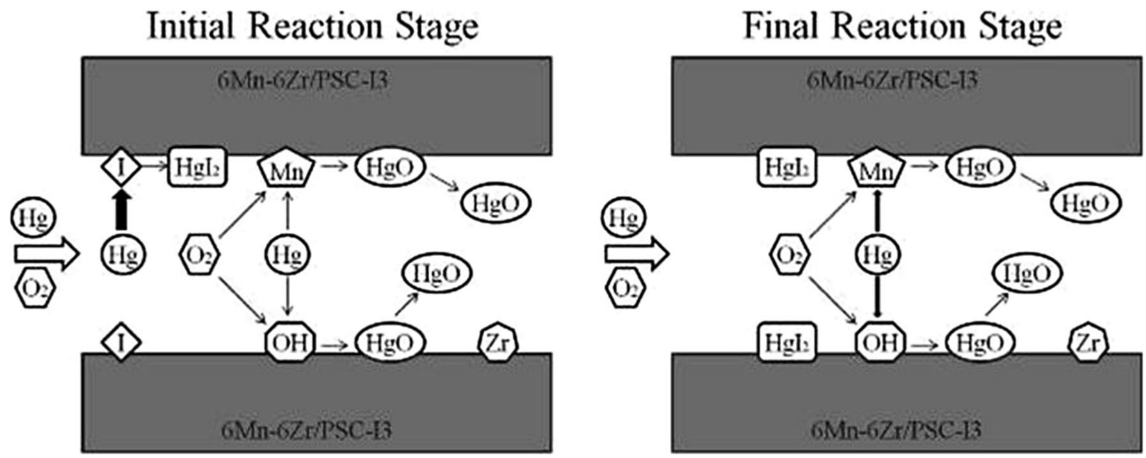

Fig. 8 Reaction mechanism of 6Mn-6Zr/PSC-I3 for Hg^0 removal at different reaction stages. At the initial reaction stage, Hg^0 is first removed by the chemical adsorption sites of C-I groups. And the Hg^0 oxidation caused by the hydroxyl (OH) oxygen and lattice oxygen played a key role at the final reaction stage (reproduced with permission from Zeng et al. 2017)

change pore structure, are also employed to improve the Hg^0 removal performance of sorbents derived from biomass char (Xu et al. 2016a; Li et al. 2015d; Lopez-Anton et al. 2015; Niu et al. 2017; Wang et al. 2018). Xu et al. (2016a) modified bamboo char (BC) using an oxidizing agent (HNO_3) and showed that the modification by HNO_3 increased the Hg^0 capture efficiency from the flue gas. The improvement was ascribed to the oxygen functional groups (such as carboxylate, carboxyl, and carbonyl groups) on the modified bamboo. In addition, the presence of water vapor improved the Hg^0 removal performance. Li et al. (2015d) modified pyrolyzed char from waste tire by H_2SO_4 and HNO_3, respectively. The results showed that the raw pyrolyzed char (T6) exhibited superior Hg^0 removal performance compared with those of acid-modified char (T6 N and T6S), attributable to the loss of sulfide functional groups on the modified samples.

Lopez-Anton et al. (2015) developed a low-cost sorbent based on leather industry waste by KOH activation and showed that the modified samples achieved the highest Hg^0 removal capacity under the N_2/O_2 atmosphere. Niu et al. (2017) treated corn stalk samples by the dielectric barrier discharge (DBD) plasma method under $N_2/O_2/H_2O$ atmosphere and found the DBD plasma-treated corn stalk sorbents to have a higher Hg^0 removal capacity compared with that of a raw corn stalk. The XPS analysis indicated that oxygen-containing functional groups increased significantly on the surface of the samples after the dielectric barrier discharge (DBD) plasma treatment, which played an important role in the removal of Hg^0. Wang et al. (2018) treated six straw chars by Cl_2 non-thermal plasma method and found that the Hg^0 removal efficiency increased from 10% to over 80% after the treatment. For example, as shown in Table 4, the Hg^0 removal capacity of T6Cl was more than 36 times than that of T6 (tobacco straw). The improved results could be ascribed to the generated C–Cl groups on the samples, which served as activated sites for Hg^0 removal.

Fly ash-based sorbents

Many investigators (Wang et al. 2016b; Hower et al. 2010) have identified fly ash (FA), a by-product of coal combustion as a promising alternative to activated carbon (AC) due to its very low cost and abundance. Related studies indicated that the fly ash has the ability to oxidize and adsorb Hg^0 in flue gas because of the presence of some oxides such as CaO, TiO_2, Fe_2O_3, CuO, Al_2O_3, and unburned carbon as part of its composition (Borderieux et al. 2004; Guo et al. 2010; Dunham et al. 2003). However, compared with activated carbon (AC), the Hg^0 removal performance of the fly ash is relatively poor (Cao et al. 2009) and requires some physical and chemical modification methods including the use of halogens and metal oxides to improve its capacity for Hg^0 (Zhao et al. 2010; Bisson et al. 2013). The modification conditions and Hg^0 removal capacities of raw fly ash and modified sorbents are summarized in Table 5.

It is well known that the compositions of fly ash played an important role in Hg^0 removal (Wang et al. 2016a; Yang et al. 2016e, 2017a, c). Wang et al. (2016a) investigated the Hg^0 removal mechanism and performance of fly ash, and they found that the fly ash had a 60% Hg^0 removal efficiency in simulated flue gas and that the presence of TiO_2, Fe_2O_3, and Al_2O_3 provided better improvement in performance compared to CaO and MgO and Al_2O_3. Furthermore, it was demonstrated that the reaction process of heterogeneous oxidation on fly ash followed an Eley–Rideal mechanism, with Fe_2O_3 considered as one of the active components on fly ash for Hg^0 removal (Yang et al. 2017a). Yang et al. (2016e, 2017c) reported Hg^0 removal at 100 °C of 89.5% for Fe_2O_3

Table 5 Reaction conditions and Hg^0 removal performance of raw fly ash and modified sorbents

Raw sorbents	Name of modified sorbents	Simulated flue gas	RT* (°C)	MRE** (%)	AC*** (μg/g)	References
Fly ash	–	$NO/HCl/SO_2/O_2/CO_2/N_2/Hg^0$	–	60	–	Wang et al. (2016a)
Fly ash	ZJM-HF	$N_2/O_2/Hg^0$	250	50	–	Yang et al. (2017a)
Fly ash	–	$N_2/CO_2/O_2/HCl/Hg^0$	100	89.5	–	Yang et al. (2016e, 2017c)
Fly ash	–	Air/Hg^0	–	–	4.5 ng/mg > 2.7 ng/mg 3.5 ng/mg 4.0 ng/mg	Zhang et al. (2017c)
Fly ash	A-HBr A-$CaCl_2$ A-$CaBr_2$	Air/Hg^0	150	98.4 67.5 46.4	–	Zhang et al. (2014b)
Fly ash	–	Air/Hg^0	–	–	100	Song et al. (2014)
Fly ash	–	Air/Hg^0	150	44	–	Zhang et al. (2015f)
Fly ash	–	Air/Hg^0	150	66.1	–	Zhang et al. (2017d)
Fly ash	KCl-FA (Fly ash) KBr-FA (Fly ash) KI-FA (Fly ash)	$O_2/CO_2/N_2/SO_2/HCl/Hg^0$	120	> 50 > 70 > 90	–	Li et al. (2013b)
Fly ash	$CuCl_2$-FA (Fly ash) $FeCl_3$-FA (Fly ash) $CuBr_2$-FA (Fly ash)	$N_2/O_2/SO_2/HCl/Hg^0$	100	> 95 > 70 100	–	Xu et al. (2013)
Fly ash	$CuCl_2$-MF (Fly ash)	$N_2/O_2/CO_2/HCl/SO_2/NO/Hg^0$	150	90.6	–	Yang et al. (2016b, c, d)
Fly ash	Mn(2)-Fe(3)-FA	$Air/N_2/Hg^0$	120	98	–	Xing et al. (2012)
Fly ash	Co/FA (Fly ash)	$Air/N_2/Hg^0$	80	76	–	Xu et al. (2014b)

*Reaction temperature

**Hg^0 removal efficiency

***Adsorption capacity

and investigated the Hg^0 reaction mechanism on its surface in the presence of HCl, suggesting that the main reaction process was in accordance with: $Hg^0 \rightarrow FeHgCl(s) \rightarrow HgCl_2$.

Halogens modification is considered to be an effective method to enhance the adsorption and oxidation of Hg^0 (Zhang et al. 2017c). Zhang et al. (2014b) compared three different halogenated fly ashes in an entrained flow reactor and found that the fly ash modified by HBr exhibited better Hg^0 removal ability as compared with metal halogens such as $CaCl_2$ and $CaBr_2$. Song et al. (2014) and Zhang et al. (2015f) also studied the Hg^0 removal performance of HBr-modified fly ash in a fixed-bed reactor and an entrained flow reactor, respectively, and found significant improvement over unmodified fly ash. Zhang et al. (2017d) further investigated the effect of NO on Hg^0 removal of HBr-modified fly ash in an entrained flow reactor and demonstrated that the introduction of NO improved the Hg^0 removal performance of the fly ash, as a result of the reaction of NO and HBr in the presence of O_2. Li et al. (2013b) developed some halogen-modified fly ash by an impregnation method and found that compared to bromine and chlorine, the iodine-modified fly ash exhibited better Hg^0 removal performance. It has been shown that both the metal ions and halogen ions contained in metal halogens acted as active sites and improved the Hg^0 removal performance (Xu et al. 2013; Yang et al. 2016b, c, d). Xu et al. (2013) suggested that metal halogens, such as $CuBr_2$, $CuCl_2$, and $FeCl_3$ loaded on fly ash, promoted the removal of Hg^0 from flue gas due to the positive role played by Cu^{2+} and Fe^{3+} cations. Yang et al. (2016b, c, d) developed a novel magnetic catalyst ($CuCl_2$-MF) based on $CuCl_2$ modified fly ash and found that the fly ash modified by 6% $CuCl_2$ achieved 90.6% Hg^0 removal from flue gas at 150 °C. In addition, when HCl was introduced into the flue gas, the $CuCl_2$-MF catalyst exhibited an excellent resistance to SO_2 poisoning. XPS and EPR analyses suggested that Cu and Cl adsorption sites were involved in the Hg^0 removal process. As shown in Fig. 9, the reaction between $CuCl_2$ and Hg^0 appears cyclical in the presence of HCl and O_2. In addition, the regeneration performance of $CuCl_2$-MF catalyst was also studied. The results of this study indicated that the regenerated catalyst showed a relatively higher Hg^0 removal capacity after thermal desorption and restoration of HCl and O_2.

In recent years, some metal oxides (e.g., manganese oxides, cobalt oxides, and iron oxides) have been used to

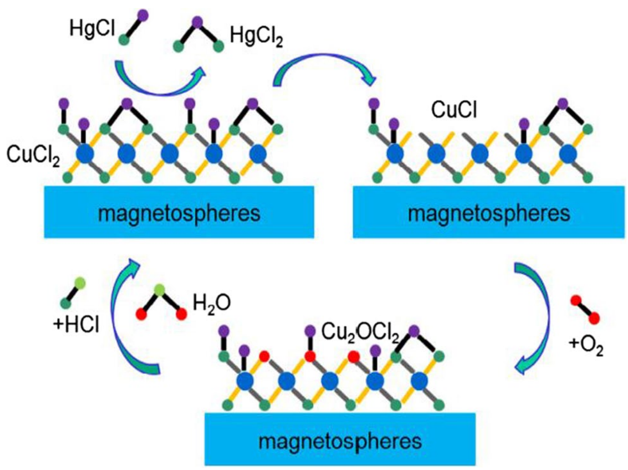

Fig. 9 Reaction process for Hg^0 removal over $CuCl_2$-MF sample in the presence of HCl and/or O_2. The Hg^0 removal over $CuCl_2$-MF samples is attributed to the synergistic role of both Cu and Cl atoms in $CuCl_2$, and the reaction between $CuCl_2$ and Hg^0 appears cyclical in the presence of HCl and O_2 (reproduced with permission from Yang et al. 2016b, c, d)

modify fly ash (FA) before its use to remove Hg^0 from flue gas. Xing et al. (2012) modified fly ash by manganese oxides and iron oxides and found that modification with Mn and Fe increased the Hg^0 removal efficiency. In particular, the Mn(2)-Fe(3)-FA samples exhibited the highest Hg^0 removal efficiency compared with raw fly ash in the presence of O_2. The XPS analysis indicated that the Mn^{4+} and Fe^{3+}, which served as active sites, could react with absorbed Hg^0 to form HgO, thereby promoting the Hg^0 removal. Xu et al. (2014b) synthesized Co-modified fly ash by a wet impregnation method and found that the sample impregnated with 9 wt% Co was very effective in Hg^0 capture, attributable to the presence of Co_3O_4 and its reaction with Hg^0 to form mercury oxides as shown in Fig. 10.

Mineral material-based sorbents

Mineral material-based sorbents have been widely studied for the treatment of Hg^0 removal in flue gas due to its low prices, abundance, and environmentally benign nature. However, various nature mineral sorbents such as zeolites, clays, and bentonites have a relatively poor capacity for Hg^0 removal, prompting the use of some agents such as halogens, metal halogens, and metal oxides under suitable modification conditions summarized in Table 6, to improve their effectiveness.

Zeolites are regarded as promising sorbents and good alternatives to activated carbon, due to their unique framework structures, favorable cation exchange properties and low cost (Wang et al. 2015b; Du et al. 2014; Chiu et al. 2014; Qi et al. 2015; Fan et al. 2012a, b). Wang et al. (2015b) investigated some zeolite sorbents for Hg^0 removal performance and demonstrated an efficiency of over 75% within 480 min at 100 °C. Du et al. (2014) developed $CuCl_2$-impregnated zeolites, and in general, found their over 80% Hg^0 removal performances were comparable to those of activated carbons. Chiu et al. (2014) further studied the effect of $CuCl_2$ modification on the physicochemical properties zeolites and their resulting effectiveness in the simultaneous removal of Hg^0, NO, and SO_2. The results of this study showed that the introduction of $CuCl_2$ decreased the pore volume and total surface area, and the $CuCl_2$-modified samples exhibited higher Hg^0 removal performances compared with their unmodified equivalents under both simulated flue gas and N_2 atmospheres. Qi et al. (2015) investigated the performance of $FeCl_3$-modified zeolites ($FeCl_3$-HZSM-5) and demonstrated that the improved Hg^0 capture efficiency obtained was due to higher surface areas and the surface-generated active Cl species. Metal oxides, with strong active and thermal stabilities, have been used as modification additives to improve the Hg^0 removal capacity of sorbents. Fan et al.

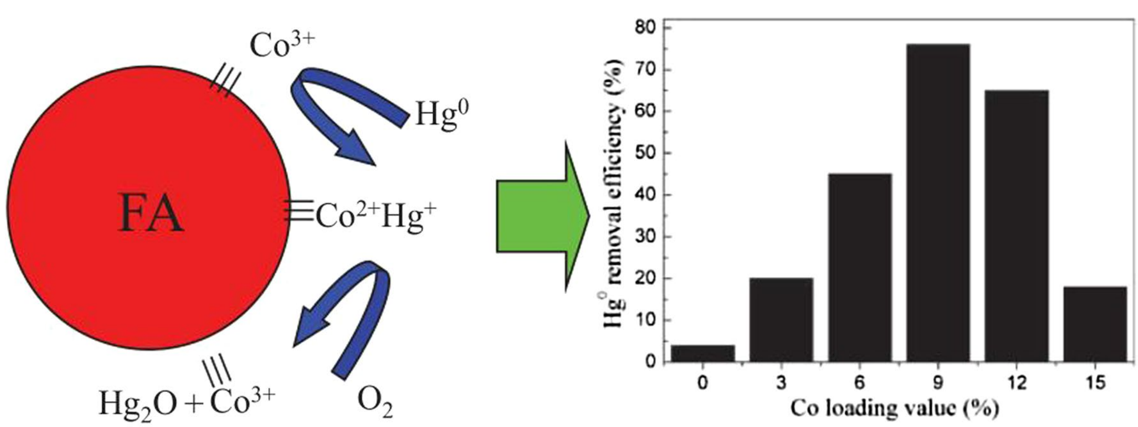

Fig. 10 Reaction mechanism of Hg^0 removal. The generated Co^{3+} on the surface of fly ash sorbents significantly promoted Hg^0 removal and O_2 played a crucial role in oxidation reactions (reproduced with permission from Xu et al. 2014b)

Table 6 Reaction conditions and Hg^0 removal performance of mineral material-based sorbents

Raw Sorbents	Name of modified sorbents	Simulated flue gas	RT* (°C)	MRE** (%)	AC*** (μg/g)	References
Zeolite	Sample G	$H_2S/H_2/CO/N_2/Hg^0$	100	> 75	–	Wang et al. (2015b)
Zeolite	$CuCl_2$-Z	$O_2/CO_2/H_2O/HCl/SO_2/NO/N_2/Hg^0$	300	> 80	–	Du et al. (2014)
Zeolite	MCM-8%	N_2/Hg^0	150	83.4	1325	Chiu et al. (2014)
		$N_2/NO/SO_2/O_2/HCl/CO_2/Hg^0$		73.4	1133	
Zeolite	$FeCl_3$-HZSM-5	$N_2/O_2/NO/HCl/SO_2/Hg^0$	120	> 95	–	Qi et al. (2015)
Zeolite	CeO_2/HZSM-5	$NO/CO_2/SO_2/H_2O/O_2/Hg^0$	200	96	–	Fan et al. (2012a)
Zeolite	Cu/HZSM-5	$NO/CO_2/SO_2/NH_3/O_2/Hg^0$	250	90	–	Fan et al. (2012b)
Clay	KBr-clay	$N_2/O_2/SO_2/H_2O/Hg^0$	180	> 57	52.96	Cai et al. (2014)
	KI-clay			> 31	487.80	
Clay	KI-Ti-PILC	$N_2/O_2/Hg^0$	180	> 65	526.32	Shen et al. (2015b)
Clay	15CeTPC	$N_2/O_2/Hg^0$	300	88.2	–	He et al. (2016a)
Clay	6Ce6MnTiP	$O_2/N_2/Hg^0$	250	72	–	He et al. (2016b)
Bentonite	Br-Ben/Na	N_2/Hg^0	140	> 90	–	Li et al. (2014b)
Bentonite	Cu-Ben	$N_2/O_2/CO_2/Hg^0$	120	> 90	–	Ding et al. (2012)
	Cl-Ben			> 45		
	I-Ben			> 90		
	Br-Ben			> 10		
Bentonite/Starch	B-S-I	$N_2/O_2/Hg^0$	120	100	604.3	Shao et al. (2016)

*Reaction temperature

**Hg^0 removal efficiency

***Adsorption capacity

(2012a, b) studied the Hg^0 removal from flue gas using both CeO_2- and CuO-modified zeolites in a laboratory-scale fixed-bed system. They found that not only did they improve Hg^0 removal compared to raw zeolite (HZSM-5), but the CeO_2/HZSM-5 and Cu/HZSM-5 also exhibited higher activities for NO removal.

Clay has been used as sorbent for Hg^0 removal due to its high abundance, good thermal stability, and layered structure. Cai et al. (2014) studied Hg^0 removal using KI- and KBr-modified clays in simulated flue gas conditions. The results indicated that the modification of KI and KBr significantly enhanced the Hg^0 removal, and the KI-modified clays had better Hg^0 removal capacity compared with KBr-modified clays. Based on these results, Cai et al. (2014) and Shen et al. (2015b) further synthesized KI-impregnated titanium-pillared clay (KI-Ti-PILC) for use to capture Hg^0 in flue gas and found that the much better performance over the raw clay was due to its more developed mesopores and higher specific surface area. Also, some metal oxides such as CeO_2 and MnO_x have been employed as modification additives due to their higher oxidation activities for Hg^0 capture (He et al. 2016a, b). He et al. (2016a) developed a CeO_2-modified pillared clay sorbent via an impregnation method, and they found that the sorbent (15CeTPC) showed a high oxidation activity of 88.2% for Hg^0 in flue gas at 300 °C in the presence of 5% O_2. He et al. further synthesized Ce-MnO_x-modified pillared clay catalysts (Ce-MnO_x/Ti-PILC), which also showed excellent Hg^0 removal performance (He et al. 2016b).

Bentonite, a type of clay mineral composed of montmorillonite, has been also used for treatment of Hg^0 in flue gas (Li et al. 2014b; Ding et al. 2012; Shao et al. 2016). Li et al. (2014b) synthesized the ammonium bromide-modified bentonite via an impregnation method and found that the modification enhanced the Hg^0 removal efficiency. Ding et al. (2012) also synthesized a number of bentonite-based sorbents modified by $CuCl_2$, $NaClO_3$, KBr, or KI, and reported that the KI-modified and $CuCl_2$-modified samples achieved better performance of about 90% Hg^0 removal at 120 °C. Furthermore, Shao et al. (2016) synthesized a novel KI-modified bentonite-starch sorbent (B-S-I) and found it to be more effective for Hg^0 removal than that of KI-modified bentonite sorbent (B-I). It was suggested that the starch–iodine complex formed by the reaction of iodine and starch promoted Hg^0 removal via the ability to release I_2, which could react with Hg^0 to form iodated mercuric compounds, thus promoting Hg^0 removal, as shown in the reaction mechanism depicted in Fig. 11.

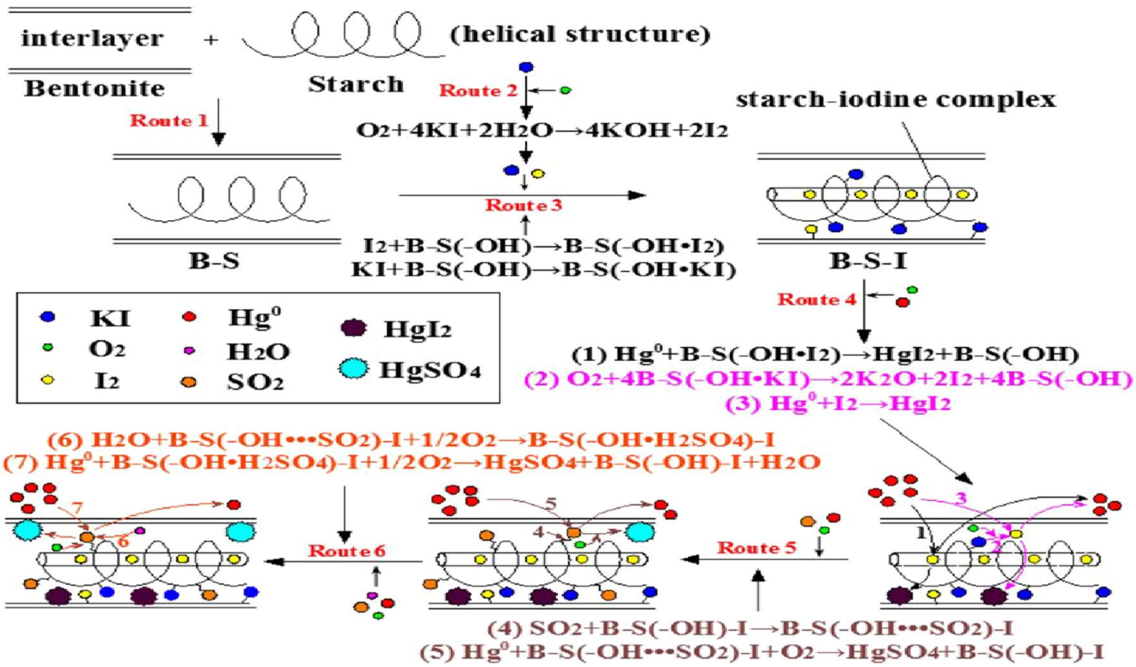

Fig. 11 Reaction mechanism of Hg^0 removal by B-S-I. The starch-iodine complex formed by the reaction of iodine and starch promoted Hg^0 removal via the ability to release I_2, which could react with Hg^0 to form iodated mercuric compounds, thus promoted Hg^0 removal (reproduced with permission from Shao et al. 2016)

Other novel Hg^0 removal technologies

In addition to the catalysts and sorbents extensively discussed above, other novel capture processes for Hg^0 in flue gas systems involving photocatalysis, plasma catalytic oxidation, and microwave catalytic oxidation under various modification conditions have been developed as attractive alternatives to conventional technologies, and are summarized in Table 7 (Wu et al. 2015a; Zhuang et al. 2014; Zhang et al. 2016a; An et al. 2016; Yang et al. 2012a, b; Liu et al. 2015a; Wei et al. 2015a, b).

Table 7 Reaction conditions and Hg^0 removal performance of novel removal methods

Novel removal methods	Modification reagents	Name of modified sorbents	Simulated flue gas	RT* (°C)	MRE** (%)	References
Photocatalysis	$Ti(SO_4)_2$	TiO_2	Air/Hg^0	55	82.75	Wu et al. (2015a)
	TiO_2	CTNTs	$N_2/O_2/Hg^0$	20	> 90	Zhuang et al. (2014)
	$Bi(NO_3)_3$/KCl	BiOCl	$N_2/O_2/CO_2/Hg^0$	–	50	Zhang et al. (2016a)
	$Bi(NO_3)_3$/KBr	BiOBr			90	
	$Bi(NO_3)_3$/KI	BiOI			> 95	
Plasma	Plasma/O_2	–	$N_2/O_2/SO_2$/NO/HCl/Hg^0	110	99.1	An et al. (2016)
	$TiCl_4$	TiO_2-B	$N_2/H_2O/Hg^0$	140	98.4	Yang et al. (2012a, b)
	$TiCl_4$	TiO_2-B	N_2/O_2/HCl/Hg^0	140	94	
	TiO_2	TiO_2	N_2/O_2/HCl/Hg^0	25	71	Liu et al. (2015a)
	SiO_2	SiO_2			15	
	$TiO_2/Mn(NO_3)_2$	Mn/TiO_2			> 90	
	$SiO_2/Mn(NO_3)_2$	Mn/SiO_2			> 80	
Microwave	$Mn(NO_3)_2$	Mn/γ-Al_2O_3	Air/O_3/Hg^0	–	92.2	Wei et al. (2015a)
	$Mn(NO_3)_2$	Mn/zeolite	Air/O_3/Hg^0	> 90	> 92	Wei et al. (2015b)

*Reaction temperature

**Hg^0 removal efficiency

Photocatalytic oxidation has been considered as a promising technology to remove Hg^0 in flue gas because it is a green process with superior oxidation ability (Wu et al. 2015a; Zhuang et al. 2014; Zhang et al. 2016a). Wu et al. (2015a) synthesized TiO_2 hollow sphere via a hydrothermal method and evaluated its performance for Hg^0 in flue gas under the irradiation of ultraviolet lamp. The results indicated that the sample showed an excellent photocatalytic oxidation for Hg^0 oxidation with a conversion of 82.75%. Zhuang et al. (2014) developed carbon-modified TiO_2 nanotubes by a hydrothermal method, which achieved an effective Hg^0 removal performance under the white light LED lamp irradiation. Zhang et al. (2016a) synthesized some BiOX (X denotes Cl, Br, and I) photocatalysts via a sample coprecipitation method for use to capture Hg^0 in flue under fluorescent light. The results of the study indicated that compared with BiOCl, BiOBr, BiOI exhibited much better Hg^0 removal capacity. It was suggested that the presence of a hole (h^+) and ion ($\cdot O^{2-}$) played key roles in BiOBr reaction system, while for BiOI reaction system, the generated I_2 might be the main species for Hg^0 oxidation.

The plasma catalytic oxidation technology has obtained much attention due to its ability to oxidize Hg^0 via the generation of active species such as O_3, OH, HO_2, and O (An et al. 2016). Yang et al. (2012a, b) studied the oxidation of Hg^0 using TiO_2 power via non-thermal plasma coupled with photocatalysis and found that the combined plasma-photocatalysis system resulted in a synergistic effect, promoting improved Hg^0 oxidation performance. Liu et al. (2015a) investigated the Hg^0 removal performance of SiO_2, TiO_2, and SiO_2 or TiO_2 supported transition metal oxide catalysts at low temperatures using a plasma-catalyst reactor. The results showed that while the non-thermal plasma could effectively enhance the Hg^0 oxidation, the presence of Mn/TiO_2 catalysts resulted in the highest Hg^0 removal efficiency of about 99% under a SED of 2.3 ± 0.3 J/L.

Wei et al. (2015a, b) synthesized Mn/γ-Al_2O_3 and Mn/zeolite catalysts via an incipient wetness impregnation method for microwave catalytic oxidation of Hg^0 in flue gas under the integrated ozone atmosphere. They reported more than 90% Hg^0 removal efficiency in the integrated microwave and ozone system, and also attributed the higher efficiency to the presence of ozone and large amounts of free radicals (O, HO_2, and OH) and their strong ability to oxidize Hg^0.

Proposed mechanism for the heterogeneous oxidation of elemental mercury

Typically, the Hg^0 can be oxidized to Hg^{2+} by the heterogeneous reactions or/and homogeneous reactions. The mechanistic aspects of Hg^0 removal using sorbents and catalysts have been extensively studied by numerous investigators (Zhao et al. 2015d; Chen et al. 2016; Zhang et al. 2017d; Xu et al. 2014b). It is well known that sorbents and catalysts promote heterogeneous reactions, which are faster reaction rate than homogeneous reactions (Presto and Granite 2006). The Deacon process, Eley–Rideal, Langmuir–Hinshelwood, and Mars–Maessen are among some of the mechanistic approaches, which have been employed to explain and quantify the heterogeneous oxidation of Hg^0 in flue gas.

The Deacon reaction

The mechanism assumes that the process by which Cl_2 (or Cl atom) can be generated by the reaction of HCl and O_2 or air at high temperature (e.g., 300–400 °C) as in Eq. (1) is the main pathway for Hg^0 catalytic oxidation in flue gas.

$$4HCl_{(g)} + O_{2(g)} \rightarrow 2Cl_{2(g)} + 2H_2O \quad (1)$$

The Deacon reaction could produce a large amount of Cl_2 in the presence of some sorbents and catalysts, thereby promoting Hg^0 removal (He et al. 2016a). Based on the results of Xu et al. (2014a) and Du et al. (2014), the Deacon reaction may be the main pathway for Hg^0 removal over the Cu-based sorbents and catalysts in HCl and O_2 atmosphere. Zhao et al. (2017b) suggested that the different reaction temperature ranges have significant effects on the Deacon reaction. As shown in Fig. 12, Hg^0 could be adsorbed by Mo or Ag on the surface of catalyst to form the Mo-Hg or silver amalgam at low reaction temperature and then combine with the active Cl species produced by a reaction of HCl and Ag-Mo/V-Ti to form soluble and adsorbable $HgCl_2$, namely the Semi-Deacon reaction. When the reaction temperature is in the range of 350–450 °C, the generated Cl_2 could begin to react with the gaseous Hg^0 to form $HgCl_2$, namely the Full Deacon reaction. Chen et al. (2014) by employing the Deacon mechanism explained that the gaseous O_2 was firstly adsorbed and activated by Ru_{cus} to generate the active O species, and then the produced active O species reacted with HCl to generate Cl_2. The reaction pathways can be described as follows:

$$2Ru_{cus} + O_2 \rightarrow 2Ru_{cus} - O^* \quad (2)$$

$$Ru_{cus} - O^* + HCl \rightarrow Ru_{cus} - OH - Cl^* \quad (3)$$

$$4Ru_{cus} - OH - Cl^* \rightarrow 4Ru_{cus} - Cl^* + 2H_2O + O_2 \quad (4)$$

$$2Ru_{cus} - Cl^* \rightarrow 2Ru_{cus} + Cl_2 \quad (5)$$

$$2Ru_{cus} - Cl^* + Hg^0 \rightarrow 2Ru_{cus} + HgCl_2 \quad (6)$$

The Eley–Rideal mechanism

In general, this mechanism assumes surface reaction involving physically adsorbed reactant (A) and chemisorbed reactant (B)

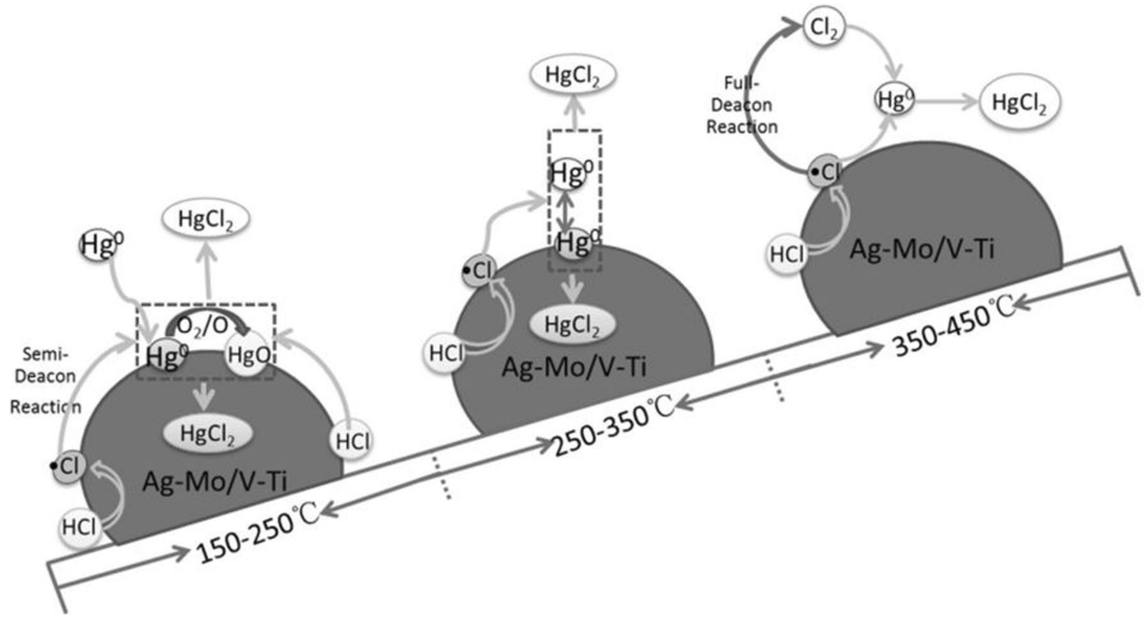

Fig. 12 Reaction mechanism for Hg^0 removal over Ag-Mo/V-Ti at different reaction temperatures. The Mo-Hg or silver amalgam formed by the reaction of adsorbed Hg^0 and Mo or Ag can react with active Cl to produce $HgCl_2$ at low reaction temperature, namely, the Semi-Deacon reaction. When the reaction temperature is in the range of 350–450 °C, the generated Cl_2 could begin to react with the gaseous Hg^0 to form $HgCl_2$, namely the Full Deacon reaction (reproduced with permission from Zhao et al. 2017b)

or reactant (A) in gas phase and chemisorbed reactant (B), and vice versa. That is, the adsorbed active species, such as HCl, could react with the gas-phase or weakly adsorbed Hg^0 to form Hg^{2+} (Zhao et al. 2014; Wang et al. 2016a). The Eley–Rideal reaction mechanism has been employed in the study of Hg^0 oxidation over selective catalytic reduction (SCR) catalysts in the presence of HCl (Yang et al. 2017d; Zhang et al. 2015d). It has been suggested that in the Hg^0 oxidation over SCR catalysts, the HCl was firstly adsorbed on the surface of the catalyst to generate active Cl sites, which could react with the gas-phase or weakly adsorbed Hg^0 to produce $HgCl_2$ (Wang et al. 2013). The specific reaction mechanism can be described as follows:

$$4HCl_{(g)} + O_2 \rightarrow 2H_2O + 4Cl^*_{(ad)} \quad (7)$$

$$Hg^0_{(g)} + Cl^*_{(ad)} \rightarrow HgCl_{(ad)} \quad (8)$$

$$HgCl_{(ad)} + Cl^*_{(ad)} \rightarrow HgCl_{2(ad)} \quad (9)$$

$$HgCl_{2(ad)} \rightarrow HgCl_{2(g)} \quad (10)$$

Similarly, the reaction of H_2S and Hg^0 also followed the Eley–Rideal reaction mechanism (Zhou et al. 2013). Related results (Hou et al. 2014a; Li et al. 2014a; Han et al. 2016; Yue et al. 2015) suggested that H_2S could be oxidized by some active species to form adsorbed active sulfur species (S_{ad}), which could further reacts with the gas-phase Hg^0 to generate HgS. The reaction mechanism can be described as follows:

$$H_2S_{(g)} + O^* \rightarrow S_{(ad)} + H_2O \quad (11)$$

$$S_{(ad)} + Hg^0 \rightarrow HgS \quad (12)$$

The Langmuir–Hinshelwood mechanism

Langmuir–Hinshelwood (L–H) mechanism (also known as Langmuir–Hinshelwood–Hougen–Watson (LHHW) in chemical reaction engineering) generally employs Langmuir's adsorption isotherm for chemisorption and assumes equilibrium adsorption and that the surface reaction is controlling. It has been used extensively to describe the bimolecular reaction between two species adsorbed on the surface of sorbents and catalysts (Zhao et al. 2016a; Liu et al. 2016a). Based on this reaction mechanism, the adsorbed Hg^0 could react with some adsorbed oxidant species, such as HBr and HCl (Lim and Wilcox 2013; Song et al. 2014). The results of prior studies indicate that the Hg^0 oxidation on the surface of some metal oxides-based sorbents and catalysts followed the Langmuir–Hinshelwood mechanism (Zhang et al. 2017b; Hou et al. 2014b; Huang et al. 2016). Jampaiah et al. (2015) and Wang et al. (2014) suggested that the Hg^0 removal on the Mn/Ce catalysts could be described by the Langmuir–Hinshelwood mechanism, whereby the adsorbed

Hg^0 could react with adsorbed active species to form Hg^{2+} as in reactions in Eqs. (13–17). Negreira and Wilcox (2013) also obtained similar results in the oxidation of Hg^0 over vanadia–titania selective catalytic reduction (SCR) catalyst. In addition, some investigators have indicated that the Hg^0 oxidation on the surface of catalyst in the presence of SO_2 could also be explained by the Langmuir–Hinshelwood mechanism and suggested that the active species derived from SO_2 could react with adsorbed Hg^0 to form $HgSO_4$ (Li et al. 2013a; Chiu et al. 2015; Zhang et al. 2016b).

$$2HCl_{(g)} + O^* \rightarrow 2Cl^*_{(ad)} + H_2O \quad (13)$$

$$Hg^0_{(g)} \rightarrow Hg^0_{(ad)} \quad (14)$$

$$Cl^*_{(ad)} + Hg^0_{(ad)} \rightarrow HgCl^*_{(ad)} \quad (15)$$

$$HgCl^*_{(ad)} + Cl^*_{(ad)} \rightarrow HgCl_{2(ad)} \quad (16)$$

$$HgCl_{2(ad)} \rightarrow HgCl_{2(g)} \quad (17)$$

The Mars–Maessen mechanism

The Mars–Maessen mechanism had been considered by numerous investigators as the most plausible mechanism for Hg^0 oxidation on the surface of metal oxide-based sorbents and catalysts (Wu et al. 2015b; Xu et al. 2016b; Qu et al. 2015). In this mechanism, the adsorbed Hg^0 could react with the lattice oxygen to form a binary mercury oxide. The Hg^0 oxidation mechanism on the Fe_2O_3–SiO_2 catalyst could be described by the reactions in Eqs. (18–22) (where M denotes Fe) (Tan et al. 2012c). Firstly, the gas-phase Hg^0 is assumed to adsorb on the surface of catalysts to form adsorbed Hg^0. Then the adsorbed Hg^0 is oxidized by the lattice oxygen from metal oxides to form HgO. Finally, the consumed lattice oxygen could be regenerated and replenished by the gas-phase oxygen from flue gas. The oxidation of Hg^0 by other metal oxides such as CuO_x, MnO_x, and CeO_2 could also be explained by the Mars–Maessen mechanism (Zeng et al. 2017; Chiu et al. 2017; Li et al. 2015e). Also, other investigators have used the Mars–Maessen mechanism to explain the oxidation of Hg^0 over some multi-metal oxide-based catalysts (Zhang et al. 2015e, 2016d; Li et al. 2016b; Zhao et al. 2016c). Zhao et al. (2015c) suggested that the Hg^0 oxidation on CeO_2–V_2O_5 catalyst surface followed the Mars–Maessen mechanism, where the synergistic effect between CeO_2 and V_2O_5 played an important role on the oxidation of Hg^0. They proposed the plausible reaction pathways as in the Eqs. (18–22), and the reaction mechanism is illustrated in Fig. 13.

$$Hg_{(g)} \rightarrow Hg_{(ad)} \quad (18)$$

$$Hg_{(ad)} + M_xO_y \rightarrow HgO_{(ad)} + M_xO_{y-1} \quad (19)$$

$$M_xO_{y-1} + 1/2O_2 \rightarrow M_xO_y \quad (20)$$

$$HgO_{(ad)} \rightarrow HgO_{(g)} \quad (21)$$

$$HgO_{(ad)} + M_xO_y \rightarrow HgM_xO_{y+1} \quad (22)$$

Summary, challenges, future research suggestions, and prospects

In this review, recent development on several catalysts and adsorbents for Hg^0 heterogeneous oxidation removal, including mainly noble metal-based catalysts, non-noble metal-based catalysts (transition metal oxides and selective catalytic reduction catalysts), activated carbon-/coke-based sorbents, biochar-based sorbents, fly ash-based sorbents, mineral material-based sorbents and other novel catalysts, are extensively discussed. Some future research suggestions and potential directions for the development of green and cost-effective technologies are summarized here.

The noble metal-based catalysts have excellent Hg^0 removal capacity and are generally regenerable and reusable to a large extent of use, but the very high costs and scarce sources greatly limited their developments and applications. Compared with noble metals, transition metal oxides and selective catalytic reduction (SCR) catalysts have several advantages such as much lower costs and more extensive sources. However, the catalytic activity for Hg^0 of transition

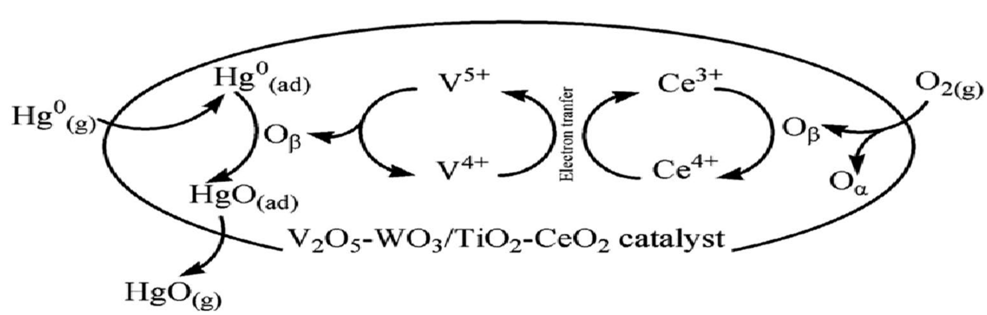

Fig. 13 Reaction mechanism of Hg^0 oxidation on CeO_2–V_2O_5 catalyst. The Hg^0 oxidation on CeO_2–V_2O_5 catalyst surface followed the Mars–Maessen mechanism, where the synergistic effect between CeO_2 and V_2O_5 played an important role on the oxidation of Hg^0) (reproduced with permission from Zhao et al. 2015c

metal oxides and selective catalytic reduction catalysts is often relatively low. Besides, their Hg^0 removal performance is greatly affected by the components of flue gas, such as halides, sulfides, vapors and alkali metal salts, and other heavy metals. The activity and stability of transition metal oxides and SCR catalysts for removing Hg^0 still need to be improved significantly using doping and other modification methods that utilize precious metals, transition metals, and nonmetals (including mixed doping and modification of multiple components). In addition, possible poisoning or deactivation of transition metal oxides and SCR catalysts by mercury itself, also needs further future investigations.

Activated carbon injection (ACI) method has been proven as one of the effective ways for Hg^0 removal from flue gas. However, the large activated carbon (AC)/Hg^0 ratio and high operation costs have limited its development. The modification with various chemical reagents (e.g., halides, sulfurs, acids, alkaline and metal oxides) can significantly improve the Hg^0 removal capacity of activated carbon, but also further increase the costs. Biochars, fly ash, and mineral materials are considered as the potential alternatives to activated carbon due to their much lower costs and more extensive sources. However, they have low Hg^0 adsorption capacity due to the poor adsorption sites on their surface. To improve the effectiveness of these adsorbents, chemical reagents are also used to modify them by increasing active sites on the surface. Unfortunately, the leaking and secondary pollution of the modified chemical reagents used over these adsorbents have greatly hindered the development and practical applications. In recent years, various advanced oxidation processes have been widely applied in the field of flue gas purification. Therefore, exploring more green and clean modification methods, such as free radical-based advanced oxidation methods, should be considered an important future priority. However, there could a limitation of terrestrial biomass (e.g., the reduction of cultivated land area and the dispersity of biomass straw). But, the ocean contains a huge biomass resource, which could be utilized. Therefore, actively exploring the utilization of marine biomass resources such as all kinds of large algae and microalgae (e.g., using marine biomass to prepare biochars and activated carbon) could provide significant resources for human development.

At present, a large number of adsorbents have been developed, but most of these sorbents lack adequate recycling and regeneration capabilities, which greatly increased the costs of application, operation, and post-processing costs and related environmental issues due to solid waste treatment problems. Developing magnetically separable and renewable sorbents should be considered as an important research direction in the future. In addition, it is reported that most of the magnetic adsorbents are still difficult to be completely separated from magnetic impurities, for example, in coal fly ash due to similar magnetic properties. Therefore, in order to completely separate the magnetic adsorbents from the magnetic impurities successfully, significant improvements in multistage magnetic field separation processes are desirable and should be pursue vigorously in future research. The separation of sorbents from fly ash can be solved by the magnetic property of sorbent materials. Therefore, magnetic properties of magnetic adsorbents could also be effectively regulated through various preparation and modification methods, and based on magnetic differences, the separation problem of adsorbents could be more effectively addressed.

In addition, other technologies such as photocatalytic oxidation, plasma catalytic oxidation, microwave catalytic oxidation, and covalent organic frameworks (COFs) adsorption oxidation developed to remove Hg^0 in flue gas have demonstrated good Hg^0 oxidation performance. However, some problems limiting process development such as high investment and operating costs, low reliability, and stability of systems/devices, low activity and anti-poisoning ability of catalysts/adsorbents and others, need to be addressed before large-scale deployment. Also, technologies utilizing catalytic or photocatalytic membrane systems should be exploited as they could remarkably reduce the demand of oxidant (by improving its retainability) and have better efficiencies for Hg^0 removal from flue gas.

Among the aforementioned catalysts, the selective catalytic reduction (SCR) catalyst is considered the most promising, with the greatest benefit of providing simultaneous removal of NO_x and Hg^0 from flue gas, and reducing investment and operating costs of existing SCR denitrification device as it could be retrofitted into its current configuration. Furthermore, research initiatives into the development of sustainable adsorbents, such biochars-based adsorbents, as potential alternatives to conventional activated carbon, should be intensified because of their very low costs and readily available renewable sources.

Conclusion

Regulatory requirements and increased public concerns regarding mercury elevation levels and persistence in the atmosphere have stimulated worldwide research efforts to develop technologies for mercury emission control. In particular, the heterogeneous catalytic oxidation and adsorption of Hg^0 from flue gas has recently been an area of major focus because of its important scientific and practical significance. The catalysts and/or adsorbents are the key to the success of the heterogeneous oxidation removal technologies for Hg^0 from flue gas. This review provides the state-of-the-art knowledge of the chemistry and the fundamental mechanistic aspects of gas–solid heterogeneous oxidation and adsorption processes for the removal of Hg^0 from the flue gas systems. It evaluates the performance and economic viability of various catalysts/sorbents for Hg^0 removal. However, this review also reveals a number of areas in which additional research are needed. These include the development of more resistant, regenerable, effective, and versatile catalysts and adsorbents; and engineering-based research such as cost–benefit analysis, techno-economic modeling and optimization of the heterogeneous catalytic and adsorptive processes for mercury removal from flue gas systems. It is hoped that this review has stimulated thinking beyond the cases presented and should spur further research needed to further the development of greener, sustainable, and more cost-effective technologies to remove Hg^0 from flue gas.

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