



Review

Possibilities of mercury removal in the dry flue gas cleaning lines of solid waste incineration units



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ABSTRACT

Dry methods of the flue gas cleaning (for HCl and SO₂ removal) are useful particularly in smaller solid waste incineration units. The amount and forms of mercury emissions depend on waste (fuel) composition, content of mercury and chlorine and on the entire process of the flue gas cleaning. In the case of high HCl/total Hg molar ratio in the flue gas, the majority (usually 70–90%) of mercury is present in the form of HgCl₂ and a smaller amount in the form of mercury vapors at higher temperatures. Removal of both main forms of mercury from the flue gas is dependent on chemical reactions and sorption processes at the temperatures below approx. 340 °C. Significant part of HgCl₂ and a small part of elemental Hg vapors can be adsorbed on fly ash and solid particle in the air pollution control (APC) processes, which are removed in dust filters. Injection of non-impregnated active carbon (AC) or activated lignite coke particles is able to remove mainly the oxidized Hg²⁺ compounds. Vapors of metallic Hg⁰ are adsorbed relatively weakly. Much better chemisorption of Hg⁰ together with higher sorbent capacity is achieved by AC-based sorbents impregnated with sulfur, alkali poly-sulfides, ferric chloride, etc. Inorganic sorbents with the same or similar chemical impregnation are also applicable for deeper Hg⁰ removal (over 85%). SCR catalysts convert part of Hg⁰ into oxidized compounds (HgO, HgCl₂, etc.) contributing to more efficient Hg removal, but excess of NH₃ has a negative effect. Both forms, elemental Hg⁰ and HgCl₂, can be converted into HgS particles by reacting with droplets/aerosol of poly-sulfides solutions/solids in flue gas. Mercury captured in the form of water insoluble HgS is more advantageous in the disposal of solid waste from APC processes. Four selected options of the dry flue gas cleaning with mercury removal are analyzed, assessed and compared (in terms of efficiency of Hg-emission reduction and costs) with wet methods and retrofits for more efficient Hg-removal. Overall mercury removal efficiencies from flue gas can attain 80–95%, depending on sorbent type/impregnation, sorbent surplus and operating conditions.

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1. Introduction

Sulfur dioxide, HCl, NO_x, dust and heavy metals including mercury belong to the most usual pollutants in the flue gas generated by solid waste incineration units (Richers, 2010; Waldner et al., 2013). The main sources of mercury in municipal solid waste (MSW) are dry cell batteries, fluorescent lamps, amalgams, old mercury thermometers and relays (Cheng and Hu, 2012). The general trend shows a clear decrease of the mercury content in MSW, however, the content of mercury in MSW can occasionally achieve significant values from the point of view of emissions from the incineration process.

The main gaseous pollutants from MSW incineration (HCl, SO₂) can be removed by wet, semi-dry or dry absorption/sorption processes (Waste to Energy, 2010), NO_x emissions are reduced by selective catalytic reduction (SCR) or selective non-catalytic reduction (SNCR) by addition of ammonia or urea to the flue gas. This article focuses on the dry flue gas cleaning methods used for the MSW incineration (Beckmann and Wen, 2011). Majority of dust from the incineration is usually removed in electrostatic filters or fabric (bag) filters. The dry methods of flue gas cleaning are advantageous particularly in smaller solid waste incineration units (Beckmann and Wen, 2011; Kong and Davidson, 2010), they usually require lower capital costs and offer simpler operation. The dry gas cleaning methods are based on an alkali sorbent injection and fabric filters for removal of solid products from the cleaning process (based on gas–solid reactions and adsorption).

Amount and forms of mercury emissions from MSW incineration depend on waste (fuel) composition, content of mercury and chlorine in waste and on the entire process of the flue gas cleaning (Nishitani et al., 1999; Jäger et al., 2006; Galbreath and Zygarlicke, 2000), i.e. flue gas composition, cooling rate of flue gas, amount and properties of dust particles, dust filters, etc. Generally, three forms of mercury can be distinguished in the flue gas: vapors of elemental Hg, vapors of chemical compounds of mercury and mercury (mainly in the form of Hg-compounds, e.g. HgCl₂, HgO, HgSO₄) adsorbed on small fly ash and APC process particles (Jäger et al., 2006). In the case of higher HCl/total mercury molar ratio and lower SO₂/HCl ratio in the flue gas (usual in MSW incineration), prevailing part of mercury (often 70–90%) is present in the form of HgCl₂ and the smaller part in the form of elemental mercury vapors (Waste to Energy, 2010; Jäger et al., 2006; Stach et al., 2005). Removal of Hg⁰ is more difficult than HgCl₂ and requires impregnated sorbents to attain its higher gas removal efficiency (over 90%).

Removal of both forms of Hg (Hg⁰ and Hg²⁺) from the flue gas depends on reactions and sorption processes at temperatures below approx. 350 °C (Stach et al., 2005; Acuna-Caro et al., 2006; Diamantopoulou et al., 2010; Ding et al., 2012; Dranga et al., 2012). The important gas phase reactant influencing conversion of Hg⁰ to Hg²⁺ is chlorine. Two reactions in flue gas play the significant role in formation and destruction of chlorine: HCl oxidation by oxygen to Cl₂ and H₂O, and reduction/destruction of chlorine by SO₂: SO₂ + 2H₂O + Cl₂ → H₂SO₄ + 2HCl. Increasing SO₂ and H₂O

concentrations together with decreasing O₂ concentration in gas will cause lower conversions of Hg⁰ to HgCl₂. Part of HgCl₂ and a small part of the elemental Hg vapors can be adsorbed on fly ash and the particles of solid sorbents removing SO₂ and HCl (e.g. active soda, calcium hydroxide, etc.). The non-impregnated active carbons, impregnated carbons (Stach et al., 2005; Diamantopoulou et al., 2010; Granite et al., 1998) or activated lignite cokes (Wirling, 2001) are able to remove preferably the mercury compounds (“oxidized forms” such as HgCl₂, HgO, etc.). Generally, vapors of metallic (elemental) Hg are adsorbed only weakly (physical adsorption) on non-impregnated AC sorbents at temperatures over approx. 150 °C. Considerably better chemisorption of elemental Hg is achieved by active carbon based sorbents impregnated with sulfur, alkali poly-sulfides, ferric chloride, CuCl₂, etc. (Cheng and Hu, 2012; Hsi et al., 2013). Also inorganic sorbents (e.g. porous mineral rocks) with similar chemical impregnation are applicable (Ding et al., 2012; Sindram and Walter, 2006). Presence of excess of ammonia in the flue gas, e.g. due to application of SNCR for de-NO_x or ammonia for de-NO_x by SCR, generally affects partition (Karf, 2010; Madsen, 2011) of forms of mercury in the flue gas (Hg/HgCl₂/HgO ratio). If application of SCR for de-NO_x of the flue gas is considered, substantial part of mercury will be oxidized and changed into compounds (HgO, HgCl₂, etc.) under such conditions (Madsen, 2011; He et al., 2009; Hong et al., 2010). The conversion to Hg²⁺ will decrease, however, with increasing NH₃ concentration (ammonia slip) in flue gas. Removal of Hg²⁺ compounds in APC processes is generally more efficient than Hg⁰ removal in both wet and dry processes of gas cleaning. Therefore any measures contributing to higher ration of Hg²⁺/Hg⁰ in flue gas will cause increasing efficiency of mercury sorption processes.

In the dry flue gas cleaning methods in MSW incineration, solid sorbents based either on soda or calcium hydroxide are used (Beckmann and Wen, 2011; Schüttenhelm et al., 2000; Nethe, 2008). The minimum temperature required for the dry sorption of acidic gases is about 130 °C and the maximum practically acceptable temperature for dry removal of HCl and SO₂ is about 340 °C. Temperatures over approx. 190 °C and 240 °C are not convenient for the powdered active carbon-based sorbents of Hg and for sulfur vapors and droplets, respectively, because of increasing risk of dust explosion and auto-ignition. The use of mineral (inorganic) sorbents of Hg without impregnation is more suitable at temperatures below approx. 180 °C. The inorganic sorbents impregnated with FeCl₃ can be deployed at maximum temperature about 280 °C (constraints given by melting and decomposition of FeCl₃). Above 280 °C only inorganic sorbents impregnated with CuCl₂ are probably suitable, however, due to catalytic effects of Cu-compounds in de-novo formation of PCDD/PCDF, their application is questionable under such conditions (Jokiluoma, 2013). Therefore, the dry sorption process for simultaneous efficient removal of acidic gas components, mercury and organic (POP) pollutants is difficult to run at temperatures over approx. 280 °C. Process option for flue gas mercury abatement depends on overall requirements on gas cleaning process, prescribed limits for air pollutants and on costs demands. For

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