Application of Multilayer Activated Carbon System for PCDD/F Removal – Regeneration of Adsorbent

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Introduction

Activated carbon injection (ACI) is effective in removing air pollutants, such as PCDD/Fs, volatile organic compounds (VOCs), polycyclic aromatic hydrocarbons (PAHs) and heavy metals (Hg in particular) from gas streams [1]. AC adsorption followed by a BF system is widely employed due to easy handling, effectiveness, and low capital cost [2]. Although ACI technology can effectively reduce PCDD/F emission from flue gases, previous study indicates that it may actually increase the total PCDD/F discharge (including that in fly ash and flue gas) from MWIs [3]. For better controlling PCDD/F emission, a multilayer AC-adsorption system (MAS) has been developed. Previous study indicates that MAS can be applied to remove vapor-phase PCDD/Fs from simulated and real flue gas with relatively high removal efficiencies [4]. PCDD/F emission is lower than 0.1 ng I-TEQ/Nm^3, the lowest emission standard of stationary source in Taiwan, even though inlet PCDD/F concentration is higher than 400 ng I-TEQ/Nm^3. MAS was further applied to simultaneously remove vapor-phase PCDD/Fs and mercury from the exhaust of small-scale pyrolysis system adopted to remediate dioxin/Hg-containing soils and extremely high efficiency in removing both toxic contaminants was obtained [5]. In this study, the replacement rate of BACs needed to ensure the compliance of PCDD/F and mercury emissions with the standards is evaluated. Moreover, the operating parameters for effective regeneration of used BACs are also evaluated.

Materials and methods

In this study, MAS was applied to simultaneously remove vapor-phase PCDD/Fs and mercury from the exhaust of small-scale pyrolysis system adopted for remediation of dioxin/Hg-containing soils. The exhaust of small-scale pyrolysis system passes through bag filter for PM removal (including solid-phase PCDD/Fs), quench tower for water vapor removal and mercury recovery, and then MAS for the removal of vapor-phase PCDD/Fs and mercury to ensure that PCDD/F and mercury emissions meet the emission limits of Taiwan. Fig. 1 shows the concept of MAS and BAC regeneration system. Commercial bead-shaped activated carbon (BAC) with average diameter of 0.72 mm, specific surface area of 1,140 m^2/g and pore diameter of 19.3 Å is used as adsorbent and a three-layer adsorption system is applied in this study. Flue gas containing PCDD/Fs and mercury is introduced from the bottom of MAS and passes through the adsorption beds. Used BACs are transferred into MAS to BAC regeneration system. The inner diameter and BAC height of each layer of MAS are 6 cm and 2.5 cm, respectively, for effective removal of high-concentration PCDD/Fs and Hg from exhaust. The MAS is operated at ambient temperature (28–32^oC). As for the operation of BAC regeneration system, three reaction times are tested.

In the MAS, vapor-phase PCDD/Fs and mercury are collected by XAD-2 resin and H_2SO_4-$KMnO_4$ solution, respectively. USEPA Method 23 is adopted for sampling and analysis of PCDD/Fs via HRGC/HRMS (Thermo Trace GC/DFS, DB-5ms with 60 m * 0.25 mm * 0.25 μm). Furthermore, USEPA Method 101A "Determination of Particulate and Gaseous Mercury Emission from Sewage Incinerators" is applied for the collection and analysis of mercury samples.

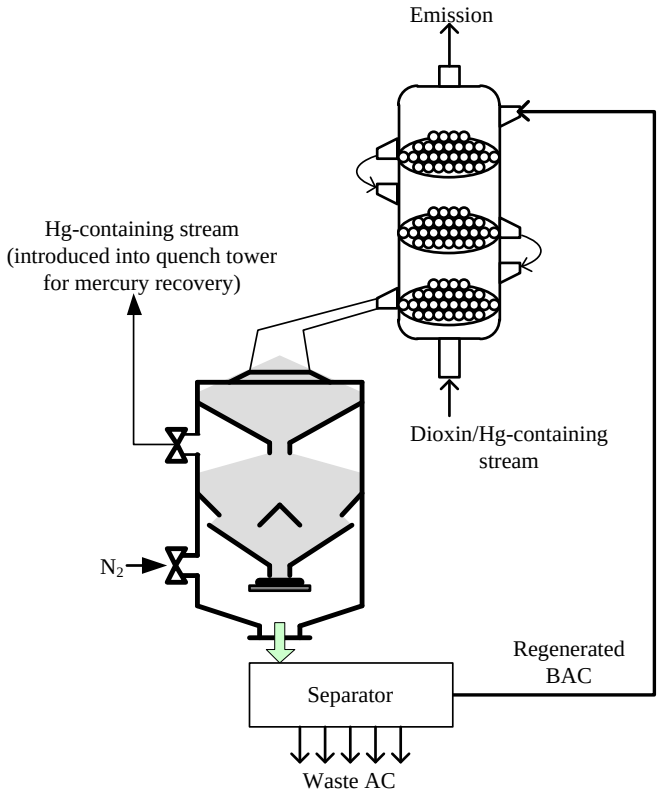

Figure 1 The concept of self-developed MAS and BAC regeneration system.

Results and discussion

Evaluation of BAC replacement of MAS

Previous study indicates that PCDD/F and mercury removal efficiencies achieved with MAS with fresh BACs as adsorbent are higher than 99.98 and 99.99%, respectively, with relatively high inlet concentrations (PCDD/Fs ≥298 ng-TEQ/Nm^3 ; mercury ≥1,290 mg/Nm^3) [5]. The emissions of PCDD/Fs and mercury are 0.0181 ng-TEQ/Nm^3 and 12.6 μg/Nm^3, respectively, which meet the most stringent emission limits of PCDD/Fs and mercury of Taiwan (0.1 ng-TEQ/Nm^3 and 50 μg/Nm^3, respectively). After long-term operation, outlet PCDD/F and mercury concentrations of each layer would increase gradually. Fig. 2 shows the PCDD/F and mercury concentrations at each layer outlet of MAS with long-term operation (27.5 hours). The results indicate that PCDD/F and mercury emissions are still lower than the emission limits of Taiwan. However, PCDD/F and mercury concentrations reach 0.1 ng-TEQ/Nm^3 and 50 μg/Nm^3, respectively, at the outlet of 2nd layer with 14 and 23 hours operation. Therefore, the replacement rate of BACs (about 18 gram/day) is calculated as 2.5 cm/day.

Evaluation of BAC regeneration system

Powder AC commonly applied to remove PCDD/Fs from real flue gas can not be recovered because it is injected into flue gas and mixed with fly ash. MAS applied in this study is installed after PM control device. BACs are mainly applied for vapor-phase pollutant removal and may be recycled. However, two coefficients including the angles of internal friction and wall friction should be considered. The angle of internal friction is the friction formed by static mechanical stress and kinetic stress between particles and particles of material tested; the angle of wall friction is the friction formed between particles of materials tested and wall of control unit assumed with stainless steel here. Lower angles of internal friction and wall friction mean that the material is relatively difficult to break during operating process. Regarding to BACs applied in this study, the angles of internal friction and wall friction are 17^o and 6.2^o, respectively. Both coefficients of BACs are lower than that of granular AC and BACs may be suitably recycled.

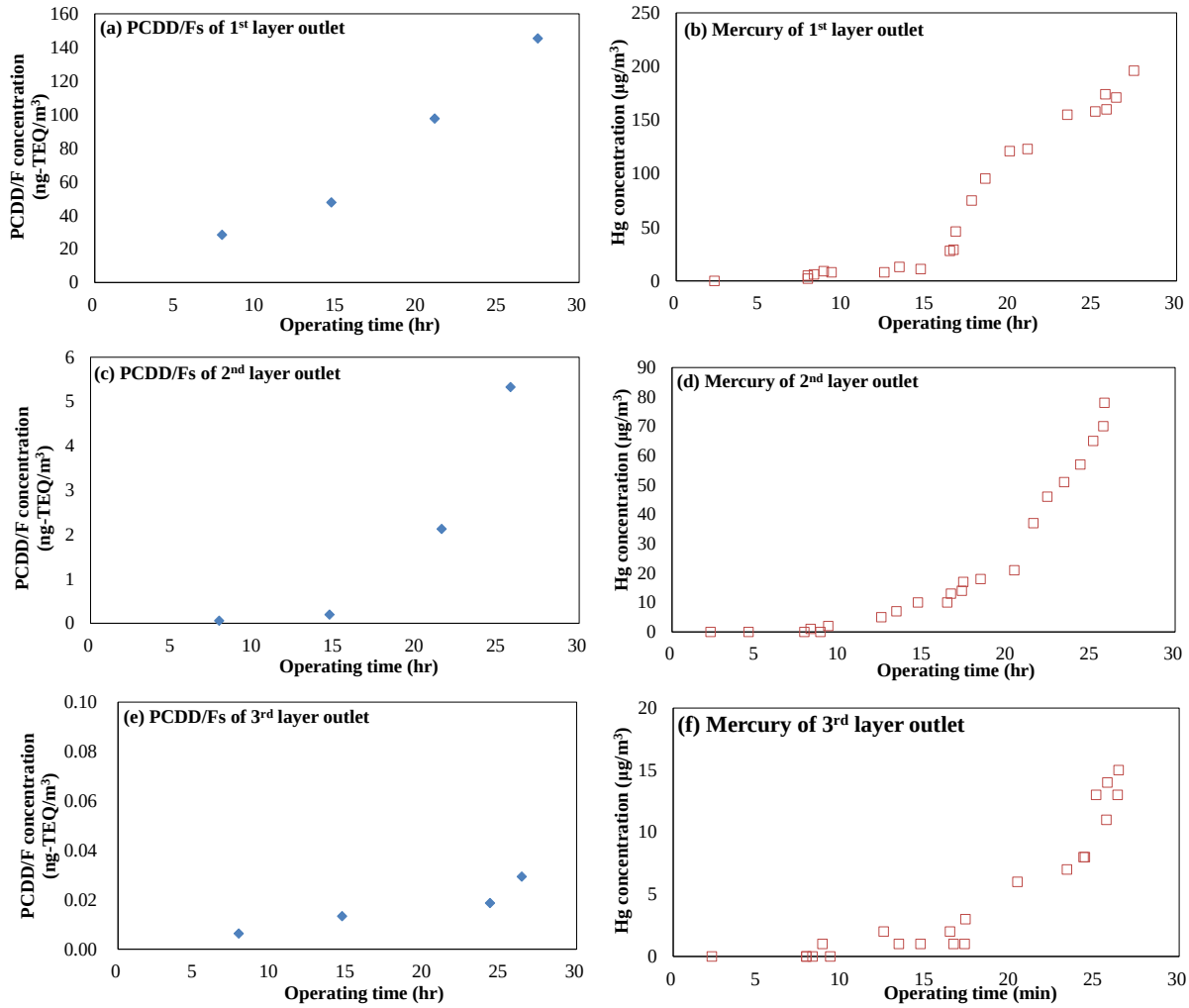

Figure 2 PCDD/F and mercury concentrations at each layer outlet of MAS with long-term operation (27.5 hours)

For BAC regeneration, temperature is controlled at 400°C and three reaction times including 1, 3 and 5 hours are tested. Nitrogen (N_2) is applied as carrier gas and pyrolysis process is applied for BAC-regenerating system. Organic compounds including PCDD/Fs adsorbed by the BACs are destroyed and mercury is desorbed from BACs to flue gas. PCDD/F and mercury concentrations of used BACs are 280,000 ng-TEQ/kg and 4,070 mg/kg, respectively. Fig. 3 shows the residual concentrations and removal efficiencies of PCDD/Fs and mercury in the BACs regenerated. PCDD/F residual concentration is lower than 500 ng-TEQ/kg with 1 hour reaction time and further decreases with increasing reaction time. The removal efficiencies of PCDD/Fs on the used BACs are ≥99%, being achieved at 400°C and ≥1 hour reaction time. Regarding to mercury, ≤36 mg/kg of residual concentration is achieved with one hour operation time and further decreases with increasing reaction time. PCDD/Fs are greatly destroyed, while mercury is efficiently desorbed to gas streams and discharged with carrier gas. The discharged flue gas is introduced into the quench tower applied to condense mercury. The specific surface area is slightly decreased from 1,140 m^2/g of fresh BACs to 939 m^2/g of regenerated BACs. This study demonstrates that MAS is effective in removing high-concentration PCDD/Fs and mercury from gas streams and reasonable replacement rate of BAC is recommended. Furthermore, used BAC can be efficiently regenerated with a pyrolysis process operating at 400°C.

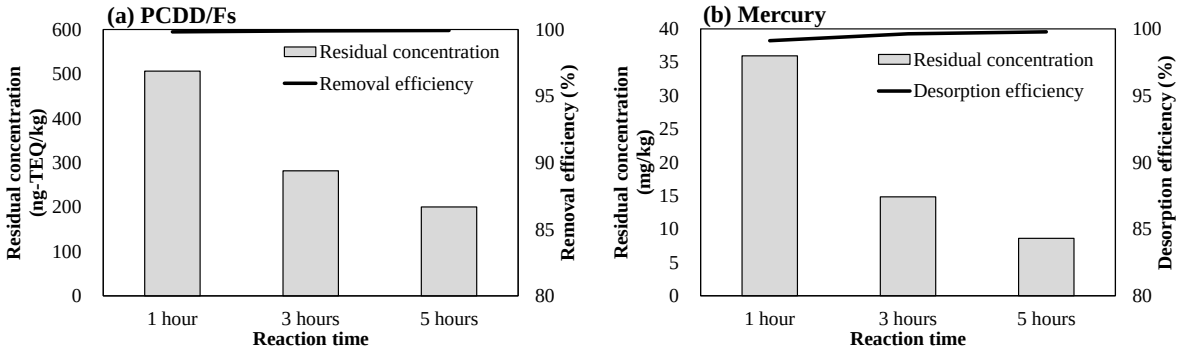

Figure 3 Residual concentrations and removal efficiencies of PCDD/Fs and mercury in regenerated BACs

Acknowledgements

The authors gratefully acknowledge the financial support provided by the National Science Council (NSC95-2221-E-008-051-MY3) and Soil and Groundwater Pollution Remediation Fund Management Board of EPA in the Republic of China.

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