CURRENT STATUS OF INCINERATION PLANTS IN JAPAN AND FLUE GAS ABATEMENT FOCUSING ON ADSORPTION TECHNOLOGY

Katsuya Kawamoto[1]

[1]National Institute for Environmental Studies, Tsukuba

Introduction

The issue of dioxins, which emerged in the last decade, has led to various changes in the disposal of solid wastes. One such change is the introduction of the gasification and melting process in conventional incineration. Abatement technologies for reducing dioxins in incineration plants have been developed in response to public concern and legal regulation (e.g. 0.1 ng-TEQ/m^3_N). There are two types of technology: one based on the principle of removal from the gas phase, and the other based on destruction mechanisms. Most plants employ the former approach, and the application of adsorbents such as activated carbon and activated coke in powder form into ducts is commonplace in Japan because it is the easiest method and does not need major alterations to the existing facility. The latter approach using catalysts is also increasing. This paper examines new processes and flue gas treatment of municipal solid waste incinerators (MSWIs) mainly in Japan. The fundamental adsorption characteristics were also investigated by experiments in order to apply the adsorption method more effectively.

Methods and Materials

Current status of gasification and melting plants: A study was conducted in 2003 to investigate the present situation of MSWIs by distributing a detailed questionnaire to about 60 gasification-melting plants. The questions concerned the performance of the facility, reduction of pollutant emissions, utility and maintenance of plant operation, and recovery of resources. The answers were analyzed, and the literature was also surveyed.

Experimental study on adsorption characteristics using a surrogate of dioxins: Experiments were conducted using a synthetic gas supply, adsorbent packed bed and sampling of the outlet gas as shown in Fig. 1. The experimental procedure used was

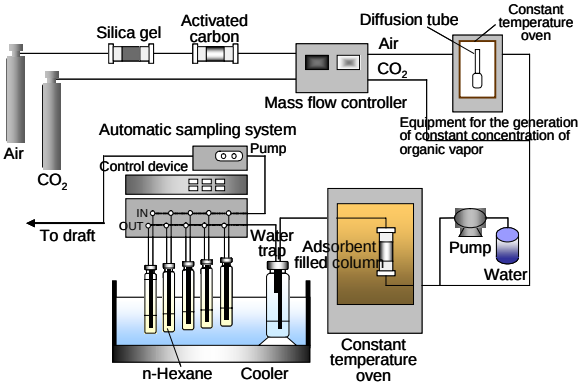


Fig.1 Experimental apparatus for adsorption test

almost the same as that described elsewhere [1], however, the number of adsorbent samples employed was increased. The adsorbents used were five kinds of activated carbon, two kinds of activated coke and an active carbide wood. The adsorption performance of the adsorbents was measured by this equipment and the results compared with reference to the physical and chemical properties. 1,2,3,4-Tetrachlorobenzene was used as a surrogate test compound in the experiment, because chlorobenzenes are known to be a possible precursor of dioxins formation and the compound was practical and suitable for the experimental equipment. The gaseous 1,2,3,4-tetrachlorobenzene in effluent gas was collected by n-hexane in a glass impinger and the concentration of the compound was measured by HRGC-LRMS.

Results and Discussions

Current status of gasification and melting plants: Gasification and melting plants have been emerging as alternative MSWIs since the dioxin issue came to the force in Japan. The change in the number of plants is shown in Fig. 2 for each type of system. The number increased to almost 60 plants in 2003 and 2004 because the period of grace of the regulation on dioxins in flue gas in MSWIs ended in December 2002. However, there are still over 1700 MSWIs in the whole of Japan, and the development of this type of incinerator is being monitored closely. The number of both fluidized bed gasification and kiln gasification plants in two years.

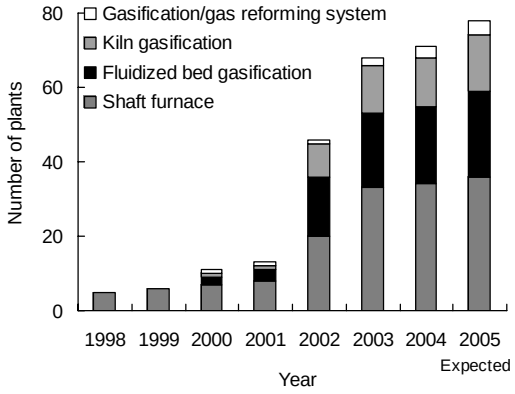
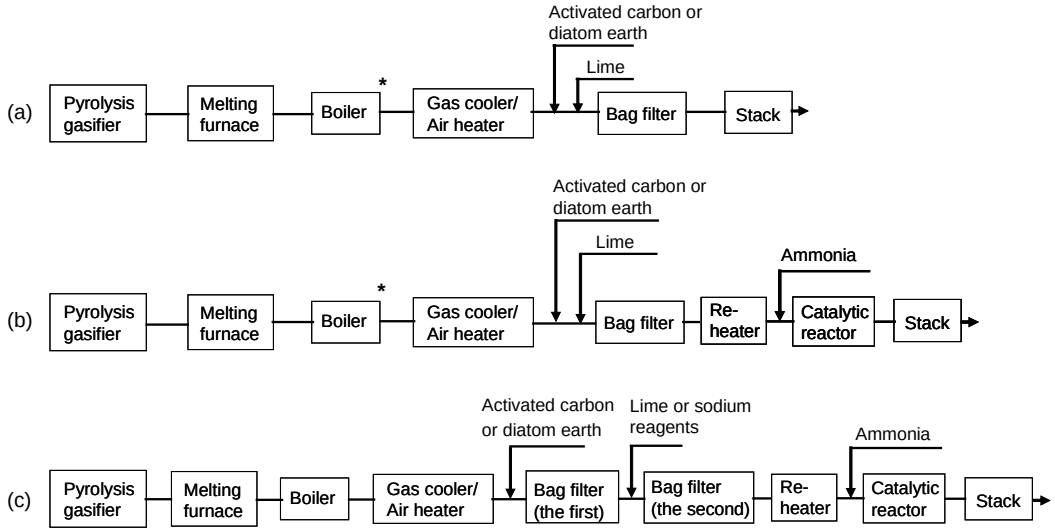


Fig. 2 Change in the number of gasification melting plants

Current status of flue gas treatment and performance for reducing pollutants: The technologies used for treating flue gas in MSWIs have remarkably changed during the last decade due to the issue of dioxins. The electrostatic precipitator (EP), which is the most general dust collection equipment, has been replaced by the bag filter (BF). This was because EPs were operated at around 300ºC at which temperature the production of dioxins was high. BF enabled operation to be conducted at low temperatures below 200ºC, and dioxins are not formed in this temperature range. Figures 3(a)-(c) show the representative patterns of flue gas treatment system employed in MSWIs including the gasification and melting process.

In principle, the dry treatment process shown in (a) is a simple process that removes acid gases and dioxins by the addition of lime and activated carbon before the bag filter. A catalytic reactor is installed into the system in order to remove nitrogen oxide and sometimes also dioxins by oxidative reaction. However, due to the independence of the catalyst on temperature, the temperature of gas often needs to be raised again higher than 200°C after low-temperature application (~170°C) of the bag filter, which may complicate the system flow from the viewpoint of energy consumption. A new process has therefore been developed as shown in (c). This process includes two series of bag filters. The first one removes fly ash and dioxins with the addition of activated carbon or another additive such as a solid reagent containing diatom earth. The second one removes acid gases with

the addition of lime. However, a new reagent containing sodium bicarbonate is beginning to be used instead of lime to improve the efficiency of removing hydrochloric acid and to solve problems in the residue caused by non-reacted lime. The fly ash collected at the first bag filter is usually put

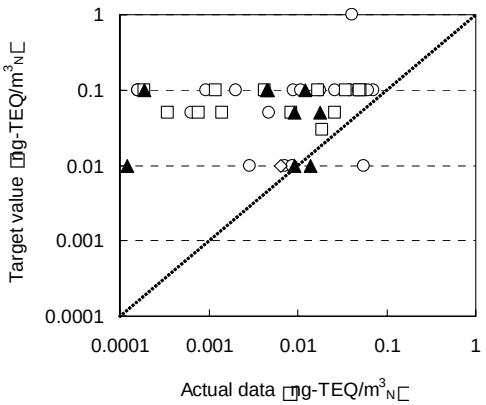


* In some cases, the boiler is not installed.

Fig.3 Representative systems of flue gas treatment employed in MSWIs

into the melting furnace and the fly ash collected at the second filter is treated properly with reagents before the final disposal. It is desirable to reduce the materials necessary to treat flue gas and to dispose of the residues. The maximum reduction of environmental pollutant load should be accomplished by the minimum use of plant utilities. In this context, the application of activated carbon adsorbents needs to be investigated.

Figure 4 shows the target value of dioxins in the stack gas and corresponding actual data measured in FY 2002 by the type of gasification melting plant. The results in most of the facilities were good because the measured data were lower than the target value. However, the opposite result was found in some facilities even though all the data were below 0.1 ng-TEQ/m^3_N, which is the standard. These findings suggest that flue



○: Shaft type furnace, □: Fluidized bed furnace, ▲: Kiln type furnace, ◊: Gas reforming type

Fig. 4 Target value and actual data of dioxins in flue gases of gasification and melting plants

gas treatment as an end-of-pipe technology generally works well. However, it is important to evaluate its efficiency for various materials used in order to ensure complete control of pollutant emissions.

Total emission of dioxins: In Japan, a technical guideline was proposed to reduce the total dioxin emissions from all streams of MSWI – emissions through air, solid and water – into the environment [2]. Subsequently, the ash melting furnace was introduced as a technical means for reducing both solid residues and dioxins. Further, this trend has been extended to the introduction of gasification melting furnaces. Data concerning the total emitted amount was not easy to calculate because it was difficult to collect precise data of the amount of solid discharges (slag and fly ash, etc.) generated and of MSW treated, and dioxins were measured only once or twice per year.

Under these limitations and uncertainties, limited data were obtained in the survey. Figure 5 shows the emissions of dioxins through each medium in a fluidized bed type gasification-melting plant. The estimated total amount of dioxins emitted from this plant was 0.53 and 1.3 μg/t-MSW in two measurement runs, both of which were very low, whereas estimations in other plants exceeded 10 μg/t-MSW. These results suggest that further work is needed to obtain reliable data on this issue. Because the amount of dioxins contained in fly ash is more than that in other substances, it is important to control *de novo* synthesis during the process of cooling the flue gas even if the melting furnace is operated at high temperature [3].

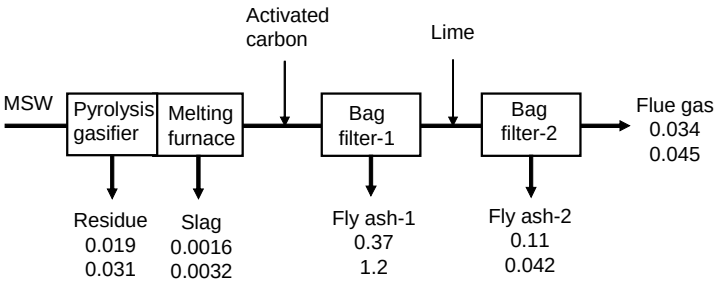


Fig.5 Emitted dioxins through each medium in a gasification-melting plant (Unit: μg/t-MSW)

Disposal of the residues: As for the residues of the pyrolysis melting plants, slag is recycled as various construction materials, and about 77% of plants answered that they perform such recycling in three types of gasification plant. In contrast, fly ash is mostly disposed of in landfill sites after proper treatment. Only one example was seen in which the fly ash was treated and transferred to a refinery.

Performance of adsorption technology as an advanced system: The number of adsorbents employed was expanded from the former (1st tier) report[1]. The three different kinds of adsorbents, namely activated carbon, activated coke and carbide wood, as recycled materials were used in the test of adsorbability. The breakthrough curves

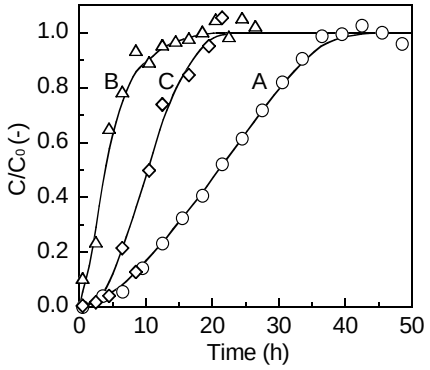


Fig. 6 Breakthrough curves of 1,2,3,4-tetra-chlorobenzene on activated carbon A, activated coke B and carbide wood C (Space velocity: 5000 h^{-1}, Moisture: 40 %, C_0: 238 $\mu g/m^3$)

taken are shown in Fig. 6. The ratio of C/C_0 increased with elapse of time. The time taken to reach adsorption saturation was short for activated coke, carbide wood and activated carbon in this order. This result might have been caused by the essential properties of the adsorbents. The specific surface areas of the materials were 285, 422 and 1213 m^2/g, respectively. The equilibrium adsorption amount of the adsorbents was calculated using the result of the breakthrough curves. Adsorption isotherms were obtained using Freundlich's formula ($Q = k\ C^{1/n}$, Q: equilibrium adsorption amount, C: equilibrium concentration of 1,2,3,4-tetra chlorobenzene). The analysis of coefficient *k* suggested that the equilibrium adsorption amount per unit amount of adsorbent at the equilibrium concentration of 1 $\mu g/m^3$ ranged from 0.2 to 4.4 mg/g-adsorbent.

Experiments were conducted for eight adsorbents, and the relationships between pore volume and equilibrium adsorption amount were obtained as shown in Figs. 7(a)-(c). The relationships suggested that the micro pore volume of 2 nm or less in diameter is the most important factor governing the adsorbability of the materials.

These findings suggest that the formulation of proper micro pore volume by an activation method using porous materials is essential for the effective removal of target pollutants. The carbide wood was expected to be a good and cost-effective adsorbent for this purpose. Further verification studies are necessary in the future.

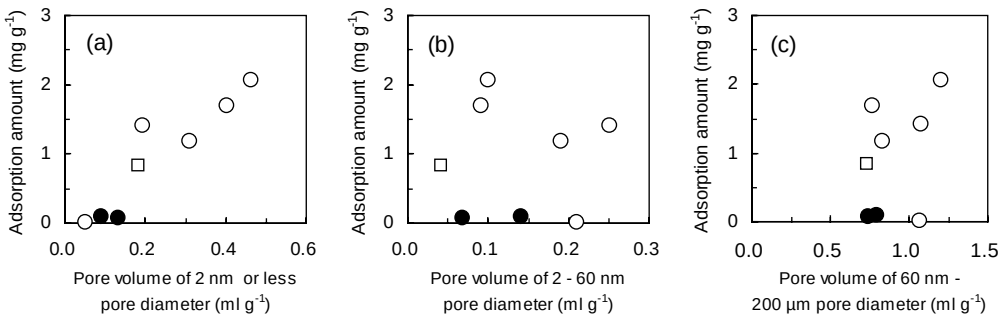


Fig. 7 Relationship between pore volume of up to 2 nm (a), 2-60 nm (b), 60nm-200 μm (c) diameter and equilibrium adsorption amount (○: activated carbon, ●: activated coke, □: wood carbide)

Acknowledgements

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