Substance Flow Approach for the Control of PCDDs/DFs
- Recent Development on Emission Control and Abatement of PCDDs/DFs in Japan

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1. Policy development on PCDDs/DFs control

The PCDDs/DFs amounts released into air from municipal solid waste (MSW) incinerators was about 1,320 g-TEQ/yr and from industrial waste incinerators was about 960 g-TEQ/yr as of November 1998[1)]. MSW incineration has 50% emission load of the total sources, which is lower compared to the load and the percentage of MSW incineration in the emission inventory reported by Hiraoka in 1990[2)]. The PCDDs/DFs amounts released from steel manufacturing (electric furnace) and sintering were 190 g-TEQ/yr (7.1% of total) and 119 g-TEQ/yr (4.5% of total), respectively[3)]. The amounts released from the zinc recovery process and aluminum alloy manufacturing process were 34 g-TEQ/yr (1.3%) and 16 g-TEQ/yr (0.6%), respectively. Environment Agency designated PCDDs/DFs as a specific substance and facilities subject to special regulations within the Air Pollution Control Law. Regulatory standards has been enforced since December 1, 1997, which specified 0.1~5 ng-TEQ/m^3 for newly established waste incineration facilities and 1~10 ng-TEQ/m^3 for existing facilities, according to their capacities. The Ministry of Health and Welfare (MHW) has carried out substantial measures for the control of PCDDs/DFs emissions in accordance with the Guideline for PCDDs/DFs Control, related to waste management, issued in 1990. The ministry chose some items, which should be dealt with by law, in the new guideline revised in 1997 and decided to regulate them by the Waste Management Law. It has enhanced design and operation standards for waste treatment facilities.

Following these regulations, the Japanese government in March 1999 arranged basic concepts on dioxin control measures which have to be taken urgently and reviewed the tolerable daily intakes (TDI) and air environmental standard. The government set a target: total PCDDs/DFs emissions in Japan would be reduced by approx. 90% compared to the amount of 1997 within 4 years. The government began to take measures such as promotion of emission reduction measures, establishment of more satisfactory measurement systems and investigation of the actual effects on human health and the environment.

2. Substance flow approach for PCDDs/DFs control

2.1 Activated carbon adsorption of emission gas and its recovery technologies

A moving bed type activated carbon adsorption tower is very effective method of removing PCDDs/DFs from emission gas. However, the treatment and safe storage of waste activated carbon is a problem to be solved. A system to recover the activated carbon and decompose adsorbed PCDDs/DFs is now under development. A demonstrative experiment was performed using an existing MSW incinerator at a 30 tons/day scale. 18 ng-TEQ/Nm^3 of PCDDs/DFs were reduced to 0.016 ng-TEQ/Nm^3 in emission gas by carbon adsorption. The activated carbon

adsorbed PCDDs/DFs was purged by N_2 and then heated in recovery tower at 400~450℃. Waste carbon was then held for 4~5 hours and cooled. PCDDs/DFs of 7 ng-TEQ/g were present in the activated carbon adsorbed were decomposed to be 0.021 ng-TEQ/g in the recovered activated carbon. The destruction rate of PCDDs/DFs in the process of activated carbon recovery is over 99%. The results of this research will be reported in the another session of this conference[4].

2.2 Treatment and recycling of combustion residues and the total PCDDs/DFs released

In 1997, Japan established a future target: the total amount of PCDDs/DFs released, including not only PCDDs/DFs contained in emission gas but also those contained in treatment residues, must be reduced to 5 μg-TEQ/ton-waste. The concept of the total release amount considering all emission matters, including not only gas-phase PCDDs/DFs but also residues, from all processes was used. Given present combustion technologies and emission gas control measures, the total emission amount would be in 100 μg-TEQ/ton-waste order on average.

The total release of PCDDs/DFs was investigated in the incinerators equipped with a residue melting furnace or a thermal dechlorination device and the gasification melting furnaces in cooperation with Japan Waste Research Foundation. 20 facilities were investigated (Table 1). When the mixture of bottom ash and fly ash was melted, the total amount released from an entire facility was between 0.29 and 11.8 μg-TEQ/ton-waste, the average was 2.30 μg-TEQ/ton-waste and the median was 0.70 μg-TEQ/ton-waste. PCDDs/DFs of about 86 μg-TEQ/ton-waste on average in residues were decomposed at a rate of over 99% and most of the released PCDDs/DFs from facilities into the environment was contained in incineration emission gas. Some facilities to treat fly ash by thermal dechlorination have begun to operate. The data of 3 cases are also shown in Table 1. As the concentration in fly ash was reduced from 0.35 ng-TEQ/g on average to 0.049 ng-TEQ/g by thermal dechlorination treatment, the total release had decreased to 3.81 μg-TEQ/ton-waste on average.

2.3 Development of gasification melting technology

Development of gasification melting technology has been promoted: waste is first decomposed by heat into pyrolysis gas, fixed carbon (char) and inorganic matter; then the inorganic matter is melted and becomes slag via the combustion heat of the pyrolysis gas and fixed carbon. This technology is characterized as follows: the oxidation of metals such as copper and iron in the process of pyrolysis is avoidable; the collection of these metals removes the catalysts of PCDDs/DFs formation; the emission gas amount can be reduced because gas is burnt with a low air ratio; and the melting slag is reusable. At present, the following three types of gasification melting furnaces have been developed: 1) Blast furnace type gasification melting; 2) External heat kiln type gasification melting; and 3) Fluidized bed type furnace gasification melting. The kiln-type gasification was demonstrated and PCDDs/DFs were measured in detail by Calaminus and Fiedler[5]. The total emission gas amounts from 9 gasification melting furnaces were investigated. The results of 5 blast type furnaces, 3 kiln type ones and a fluidized bed type are shown in Table 1. The PCDDs/DFs concentrations in melting emission gas and slag were low for all types of furnaces. The concentration in melting fly ash was 0.17 ng-TEQ/g on average. The average of the total releases was 4.15 μg-TEQ/ton-waste (0.07~9.89μg-TEQ/ton-waste).

Table 1 Residue Melting, Gasification, Thermal Dechlorination and Total Amount of PCDDs/DFs Released

		Concentration				Amount released μg TEQ/ton waste			
		No of data	Min.~ Max.	Average	Median	No of Data	Min.~ Max.	Average	Median
Melting of mixed bottom and fly ashes	Incinineration flue gas ngTEQ/m^3	8	0.026~ 1.5	0.37	0.08	8	0.15~ 10.1	2.39	0.46
	Bottom ash ngTEQ/g	6	0.00~ 0.047	0.017	0.015	6	(0.00~ 3.4)	(1.23)	(1.01)
	Fly ash ngTEQ/g	7	0.27~ 7.36	2.05	1.00	6	(11.7~ 392)	(85.4)	(28)
	Melting flue gas ngTEQ/m^3	11	0.00~ 2.27	0.46	0.19	11	0.00~ 1.39	0.21	0.019
	Slag ngTEQ/g	10	0.00~ 0.00025	0.00009	0.00008	9	0.00~ 0.02	0.0077	0.01
	Melting fly ash ngTEQ/g	11	0.0047~ 0.06	0.033	0.022	11	0.027~ 1.5	0.28	0.13
	Others (Metals) ngTEQ/g	1	-	0.0002	-	1	-	0.002	-
	Total release μg TEQ/ton waste	11	-	-	-	10	0.29~ 11.8	2.30	0.70
Gasification melting	Melting flue gas ngTEQ/m^3	9	0.0006~ 0.039	0.016	0.012	9	0.005~ 0.35	0.13	0.10
	Slag NgTEQ/g	6	0.00~ 0.00026	0.00006	0.00003	6	0.00~ 0.02	0.0055	0.0014
	Melting fly ash ngTEQ/g	9	0.002~ 0.33	0.17	0.13	9	0.05~ 9.53	4.01	2.73
	Others (nonburnables) ngTEQ/g	1	-	0.0034	-	1	-	0.055	-
	Total release μg TEQ/ton waste	9	-	-	-	9	0.07~ 9.89	4.15	2.74
Thermal dechlorination	Incineration flue gas ngTEQ/m^3	3	0.096~ 0.32	0.21	0.2	3	0.41~ 1.59	1.01	1.04
	Bottom ash NgTEQ/g	1	-	0.002	-	1	-	0.30	-
	Fly ash NgTEQ/g	2	0.29~ 0.41	0.35	0.35	2	(8.59~ 26.3)	(17.45)	(17.45)
	Waste water NgTEQ/L	1	-	0.094	-	1	0.0098	0.0098	-
	Dechlorination flue gas ngTEQ/m^3	1	0.00005	-	-	1	-	0.00005	-
	Dechlorination ash ngTEQ/g	3	0.0076~ 0.078	0.049	0.06	3	0.225~ 3.99	2.69	3.85
	Waste water NgTEQ/L	1	-	20	-	1	-	0.006	-
	Total release μg TEQ/ton waste	3	-	-	-	3	0.97~ 5.58	3.81	4.89

3. Substance flow of coplanar PCBs (Co-PCBs)

Co-PCBs can be classified into two groups according to their sources. One group is Co-PCBs contained in PCBs products manufactured in the past. These are regarded as intentional products. Another is Co-PCBs formed as a byproduct in various incineration processes including MSW incineration. These are regarded as unintentional products. We analyzed the input/output flow of Co-PCBs in the existing MSW incineration processes[6].

We examined whether MSW incineration is a process of Co-PCBs destruction or formation by comparing the input and output levels and determining whether PCDDs/DFs or Co-PCBs are the bigger contributors at the TEQ level. The amount of PCBs was 13,000~33,000 μg/ton-waste, the amount of Co-PCBs was 450~550 μg/ton-waste and the TEQ was 0.13~0.29 μg-TEQ/ton-waste. As the inflow amount of PCDDs/DFs was 1.2 μg-TEQ/ton-waste and the total inflow of PCDDs/DFs and Co-PCBs was 1.5 μg-TEQ/ton-waste, the TEQ ratio of Co-PCBs was 19%. The total release of TEQ-based Co-PCBs amount released was 2.8 μg-TEQ/ton-waste, which exceeded the input amount of 0.29 μg-TEQ/ton-waste. The contribution rates of fly ash and emission gas in the total output amount were approx. 58% and 41% respectively. On the other hand, the total output of Co-PCBs and PCBs was below the total input amount. The input of total PCBs was 13,000~33,000 μg/ton-waste, whereas the total output amount was 1,000 μg/ton-waste, thus over 90% of PCBs were decomposed. TEQ-based Co-PCBs are formed in the incineration process but Co-PCBs and PCBs as a whole are decomposed. According to the input and output amounts of each Co-PCBs congener, congeners were divided into two groups: one group the amount of which drastically increased and a second group which decreased. In particular, highly toxic #126 and #169 of non-ortho Co-PCBs increased by 10 to 50 fold during the incineration process and these Co-PCBs contributed to the rise in total TEQ-based output amount. To the contrary, #105, #114 and #118 were decomposed at a rate of over 90%. It has been suggested that non-ortho Co-PCBs are selectively formed during the incineration process. Although the formation mechanism seems to be same as that for PCDDs/DFs, further examination of this matter is necessary. This investigation was conducted for a relatively old type facility with PCDDs/DFs concentration of 1~10 ng-TEQ/Nm^3 in emission gas. Another investigation for new facilities is presently being undertaken.

References

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