

Emission Characteristics of PCDDs/PCDFs from Ferrous Metal Foundries

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Four iron-ore sintering furnaces, thirteen steel melting furnaces, and three alloy melting furnaces were selected from eighteen plants in iron-and-steel foundries. PCDDs/PCDFs samplings were achieved two times at the stacks of each furnace. The emission levels of PCDDs/PCDFs were 0.615 ng-TEQ/Sm³ in iron-ore sintering, 0.097 ng-TEQ/Sm³ steel-melting, and 0.024 ng-TEQ/Sm³ in alloy-melting processes on average. PCDFs showed an approximately four times higher concentrations than PCDDs, revealing that the ratio of PCDDs to PCDFs averaged approximately 20:80. The main contributor to total TEQs was 2,3,4,7,8-PeCDF, and its TEQ value was in the range of 44% to 52% of total TEQs. Electric furnaces have released slightly lower levels of PCDDs/PCDFs than sintering furnaces. But, the emission amounts of off-gas from electric furnace were up to about 1.5 millions standard cubic meters per hour, depending on the numbers of electric furnace in a plant. This is mainly due to sequence-batch operation of electric furnace, and thus the building is totally enclosed in order to minimize dust and gaseous emissions. Accordingly, off-gases contain very high concentration of oxygen (20.5%) and carbon monoxide (410 ppm) on average.

Key words : PCDDs/PCDFs, sintering, steel melting, alloy melting

1. Introduction

Iron-and-steel foundries can be defined as those that produce gray or white iron and steel castings, which are solid solution of iron, carbon, and various alloying materials. The castings are produced from scrap iron, pig iron, and foundry returns by melting, alloying, and molding. The major operations include: 1) raw material preparation, 2) metal sintering, 3) metal melting, and 4) casting and finishing (Fig. 1)^{1,2)}.

First, raw materials require blending prior to sintering operation. This can involve layering the materials on prepared area into the precise quantities required by sintering operation. Some flux materials can be added at this stage with recycled materials from downstream operations. Ore blend is recovered from the beds and transferred to storage pit. In the mixing stage other additives can be added to the ore blend. These additives include limestone (CaCO₃), collected dust from sintering and melting processes, and recycled

sinter in a size of 4 to 5 mm from sinter screening^{2,3)}.

In the sintering process, the blended-and-mixed fine iron ore is melted into lump iron by the heat (1,300~1,480°C) of burning coke breeze (<5 mm). A sintering furnace is operated as down-draft sintering on a continuous basis, which is a planar steel plate open at the top and equipped with traveling grate with slots at the bottom. Combustion air is introduced downwards through the layer of materials to be sintered by the induced-draft fan. Due to the coke combustion and the insufficient supplement of air, emissions released from the sintering furnaces include the high levels of particulate matter, carbon monoxide, sulfur dioxide, nitrogen oxide, and small quantities of chlorine compounds⁴⁻⁶⁾. Thus, the sintering process has been indicated as an emission source of polychlorinated dibenzo-*p*-dioxins/polychlorinated dibenzofurans (PCDDs/PCDFs)⁷⁻¹⁰⁾.

Metal melting process is accomplished primarily in a blast furnace, which is a major metal melting furnace,

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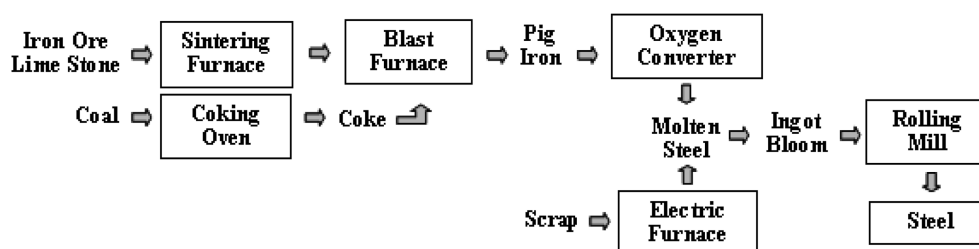


Fig. 1. Schematic diagram of integrated works in iron-and-steel foundries.

and to a lesser extent in an electric arc or induction furnace. Blast furnace is typically a vertical and cylindrical shell with refractory-lined inner wall. Sintered metal, coke and fluxes are loaded into the furnace, and pure oxygen or air is injected through tuyeres at the base of furnace to burn off the iron-bearing carbon. Heat, produced by burning coke, melts the iron which flows down and is tapped from the bottom of the furnace. Emission gas is all recycled as a fuel for power plant^{11,12}. In Korea, about 70% of all iron castings are produced using blast furnaces, while about 30% of those rely on electric arc or induction furnaces.

Scrap metal melting is performed almost at a electric arc or induction furnace in Korea. In an electric furnace, which is operated as sequence-batch basis, electric current flowing from cathode to anode through the scrap charged. Electrical resistance of the scrap causes the scrap to be heated up and molten. Molten scrap is poured into the crucible by tilting the electric furnace. Fugitive dust is controlled by enclosing a barrel furnace in a hood system and ducting the emission to a fabric filter. Principally, even though there is no combustion process in an electric furnace, the melting process for scrap metal has been also indicated as an emission source of PCDDs/PCDFs because of a partial combustion of chlorine-containing plastics or liquids¹³⁻¹⁵.

Although electrostatic precipitator or bag filter is commonly used to control emissions from the sintering and melting processes prior to releasing air pollutants into atmosphere, emissions released from these processes can include the high levels of carbon monoxide, sulfur dioxide and nitrogen oxide, and the small quantities of particulate matter, organic compounds, and chlorine and fluorine compounds. Therefore, this

study was performed to investigate emission levels of PCDDs/PCDFs in the sintering and melting processes of the iron-and-steel foundries in Korea.

2. Materials and Methods

Four iron-ore sintering furnaces, thirteen steel-melting furnaces, and three alloy-melting furnaces were selected from eighteen plants in iron-and-steel foundries. PCDDs/PCDFs samplings were achieved two times at the stacks of each furnace by using a sampling train: which consisted, in order, of a probe, a cylindrical filter assembly allowed insertion of a thermocouple (silica fiber thimble, 90 mm in length and 25 mm outside diameter, Whatman: at least 99% efficiency on 0.3 μm dioctyl phthalate smoke particles), three impingers (two of which were filled with 150 ml of distilled water, and one of which was empty), a sorbent (XAD-2) module allowed insertion of a thermocouple, and two impingers (one of which was filled with 150 ml of ethylene glycol, and the other was empty).

After spiking 2 ng of $^{37}\text{Cl}_4$ -2,3,7,8-TCDD to the sorbent, two traverse-point sampling over the respective cross sections was iso-kinetically (isokinetic factor of 95~105%) done at the stack for about 5 hours. During the sampling, the cylindrical filter was kept below 120°C with cooling water, and sorbent trap was kept below 30°C by chilling the impingers with ice water.

After sampling, the PCDDs/PCDFs samples were divided into two portions: one is solid portion (cylindrical filter and sorbent) and aqueous portion (impinger water, ethylene glycol, probe-rinsed solvent). An internal standard cocktail, 2 ng of $^{13}\text{C}_{12}$ -PCDDs/PCDFs was

added to each portion of samples as internal standards prior to extraction. Then, each portion of samples was separately extracted with toluene: Soxhlet extraction for solid portion and liquid extraction for aqueous portion. Toluene extracts were combined into one and cleaned up by 5 ml of concentrated sulfuric acid 4 or 5 times, followed by silica gel column cleanup eluting 150 ml of n-hexane, and basic alumina column cleanup eluting 100 ml of 2% dichloromethane in n-hexane (this portion was discarded) and then 150 ml of 50% dichloromethane in n-hexane. Final eluate was concentrated to a volume of 1 ml, and further concentrated to about 50 μ l after spiking 2 ng of recovery standards.

Concentrated eluates of the samples were analyzed by high resolution gas chromatograph/high resolution mass spectrometer (HRGC/HRMS). The HRGC/HRMS setup consisted of a Agilent 6890 GC coupled with Autospec Ultima (Micromass Co.) with OPUS quantification programme. Selected ion monitoring with electron impact of 36 eV was performed above a resolution of 10,000 with an SP-2331 column of 60 m \times 0.32 mm ID \times 0.25 μ m [120°C (3 min) \rightarrow 10°C \cdot min⁻¹ to 200°C (3 min) \rightarrow 3°C \cdot min⁻¹ to 265°C (15 min)]. The eluates were introduced in splitless mode with a flow rate of 2.5 ml \cdot min⁻¹ helium, and temperatures of injector and ion source were 260°C and 270°C, respectively.

Toxic equivalents (TEQs), expressed as 2,3,7,8-TeCDD values, were calculated without correction of oxygen concentration in flue-gas stream by using the international toxicity equivalency factor and OPUS quantification programme. All processes mentioned above, including sampling, sample preparation, and HRGC/HRMS analysis, were performed according to the Korean Standard Testing Method (KSTM) for Dioxins and Furans¹⁶⁾.

3. Results and Discussion

In the iron-ore sintering furnace, the volumes and temperatures of emission gases were in the range of 11,818~23,579 Sm³/min (average 18,816 Sm³/min) and 150~173°C (average 161°C), respectively. Also, off-gas

per ton of products was averaged about 2,481 Sm³/ton ranging from 2,064~3,345 Sm³/ton, and the major air pollution control device (APCD) was an electrostatic precipitator (EP).

As shown Table 1, PCDDs/PCDFs emissions were in the range of 0.333 to 1.342 ng-TEQ/Sm³, and averaged 0.615 ng-TEQ/Sm³ (standard deviation(s)= 0.321) from eight measurements. Like PCDDs/PCDFs emission pattern of municipal solid waste (MSW) incinerator¹⁷⁻¹⁹⁾. Emission gases contained four times or more PCDFs than PCDDs, showing that the ratio of PCDDs to PCDFs was 18:82 on average (Table 2). The most abundant 2,3,7,8-congener was 2,3,4,7,8-PeCDF followed by 1,2,3,7,8-PeCDD, and their TEQ values constituted 51.7% and 8.5% of total TEQs, respectively (Fig. 2). The most toxic 2,3,7,8-TCDD was detected at the level of less than 5% of total TEQs.

Currently, the major problem of sintering furnace is that it emits one million or more standard cubic meters of off-gases per hour, and releases many and much higher pollutants such as particulate matter, carbon monoxide, sulfur dioxide and nitrogen oxide into the atmosphere, including PCDDs/PCDFs. A remarkable point is the fact that off-gas released contains about 1.6% of carbon monoxide^{20,21)} and this carbon monoxide can not be controlled by electrostatic precipitator. This is mainly due to the insufficient air supplement by down-draft operation for reducing the iron ore, when coke breeze is burned as a both fuel source and reducing agent ($\text{Fe}_2\text{O}_3 + \text{C} \rightarrow 2\text{FeO} + \text{CO}$, $\text{FeO} + \text{C} \rightarrow \text{Fe} + \text{CO}$). This reducing atmosphere of sintering furnace leads to the high emissions of carbon monoxide including other pollutants. Thus, the proper treatment process, such as re-burning of flue gas, is further needed with the bag filter or electrostatic precipitator as APCDs to for reduce the carbon monoxide from the flue-gas stream of sintering furnace.

In the electric steel-melting furnaces, which were all electric arc or induction furnaces, the volumes and temperatures of emission gases were in the range of 6,181~24,673 Sm³/min (average 16,147 Sm³/min) and 38~75°C (average 57°C), respectively. Also, the off-gas per ton of product was averaged about 10,239 Sm³/ton

Table 1. PCDDs/PCDFs Emission from Ferrous Metal Manufacturing Facilities

Plant Name	Emission process	APCD	E. Gas (Sm ³ /min)	E. Gas /Prod. (Sm ³ /ton)	E. Gas Temp. (°C)	PCDDs/PCDFs (ng-TEQ/Sm ³)			
						1st	2nd	Mean	
Iron	A1	SF#1	EP	11,818	3,345	150	0.410	0.333	0.372
		SF#2	EP	23,579	2,439	151	0.594	0.574	0.584
	A2	SF#1	EP	19,989	2,076	173	1.342	0.395	0.869
		SF#2	EP	19,876	2,064	168	0.537	0.731	0.634
	Average			18,816	2,481	161	0.615		
	Steel	B1	EAF	BF	10,668	7,301	56	0.093	0.018
B2		EAF	BF	24,607	12,932	46	0.129	0.036	0.083
B3		EAF	BF	14,889	9,424	67	0.040	0.036	0.038
B4		EAF	BF	12,478	10,368	70	0.072	0.166	0.119
B5		EAF	BF	20,090	10,847	64	0.036	0.031	0.034
B6		EAF	BF	23,515	8,621	66	0.471	0.558	0.515
B7		EAF	BF	6,181	4,428	43	0.027	0.021	0.024
B8		EAF	BF	24,673	13,776	45	0.091	0.059	0.075
B9		EAF	BF	11,084	13,301	68	0.015	0.014	0.015
B10		EAF	BF	10,956	12,621	63	0.016	0.045	0.031
B11		EAF	BF	9,774	20,280	38	0.104	0.028	0.066
B12		EAF	BF	24,619	11,344	75	0.156	0.223	0.190
B13		EAF	BF	16,377	12,001	45	0.024	0.024	0.024
Average			16,147	10,239	57	0.097			
Alloy	C1	EAF	BF	652	9,551	73	0.048	0.011	0.030
	C2	EAF	BF	1,210	52,011	104	0.013	0.052	0.033
	C3	EAF	BF	886	58,365	55	0.001	0.017	0.009
	Average			916	25,863	78	0.024		

Note: SF: Sintering Furnace, EAF: Electric Arc Furnace, EP: Electrostatic Precipitator, BF: Bag Filter, E. Gas/Pro.: Emission Gas/Production,

ranging from 4,428–20,280 Sm³/ton, and the major APCD was a bag filter.

PCDDs/PCDFs emissions were in the range of 0.014 to 0.558 ng-TEQ/Sm³ (average 0.097 ng-TEQ/Sm³, $s=0.135$) from twenty six measurements. Like the sintering furnaces, PCDFs showed an approximately four times higher concentrations than PCDDs, revealing that the ratio of PCDDs to PCDFs was 22:78 on average. The most abundant 2,3,7,8-congener was also 2,3,4,7,8-PeCDF followed by 1,2,3,7,8-PeCDD, and their TEQ values constituted 44.5% and 9.7% of total TEQs, respectively. 2,3,7,8-TCDD was detected at the level of less than about 6% of total TEQs.

Electric furnaces have released slightly lower levels of PCDDs/PCDFs than sintering furnaces. But, the off-gases were up to about 1.5 millions standard cubic meters per hour, depending on the numbers of electric

furnaces inside plants, and contained a very high concentration of oxygen (20.5%) and carbon monoxide (410 ppm) on average. This is thought to be mainly due to the sequence-batch operation of electric furnace: metal scraps are charged into furnace, then electric current flows through the scrap, and after molting is completed molten iron is poured into the crucible by opening the lid and tilting electric furnace. At this time, fugitive dust is generated out of furnace, and normally captured by enclosing the barrel furnace in a canopy-hood system. But, this capturing system for fugitive dust leads to the lots of off-gas generation and a very high oxygen concentration in the flue gas stream.

In the electric alloy-melting furnaces, which were all electric arc or induction, the volumes and temperatures of emission gases were in the range of 652–1,210 Sm³/min (average 916 Sm³/min) and 55–104°C (average

Table 2. PCDDs/PCDFs emission of sintering and melting furnaces in the iron and steel foundries

2,3,7,8-PCDDs/PCDFs	PCDDs/PCDFs (ng I-TEQ/Sm ³)					
	Sintering Furnace		Electric Furnace			
	Iron		Steel		Alloy	
2,3,7,8-TCDD	0.022	3.64	0.006	5.68	0.002	7.75
1,2,3,7,8-PeCDD	0.052	8.48	0.009	9.71	0.002	8.45
1,2,3,4,7,8-HxCDD	0.007	1.14	0.001	1.11	0.000	0.70
1,2,3,6,7,8-HxCDD	0.013	2.10	0.003	2.72	0.001	2.11
1,2,3,7,8,9-HxCDD	0.010	1.55	0.002	1.58	0.000	0.70
1,2,3,4,6,7,8-HpCDD	0.005	0.75	0.001	0.75	0.000	0.70
OCDD	0.001	0.08	0.000	0.04	0.000	0.00
PCDDs	0.109	17.74%	0.021	21.59%	0.005	20.42%
2,3,7,8-TCDF	0.036	5.82	0.009	9.67	0.002	8.45
1,2,3,7,8-PeCDF	0.015	2.46	0.004	3.91	0.001	2.82
2,3,4,7,8-PeCDF	0.318	51.69	0.043	44.49	0.011	47.18
1,2,3,4,7,8-HxCDF	0.024	3.84	0.004	4.15	0.001	4.23
1,2,3,6,7,8-HxCDF	0.047	7.63	0.007	6.75	0.002	7.75
2,3,4,6,7,8-HxCDF	0.050	8.12	0.007	6.75	0.002	7.75
1,2,3,7,8,9-HxCDF	0.007	1.16	0.001	0.75	0.000	0.00
1,2,3,4,6,7,8-HpCDF	0.008	1.34	0.001	1.30	0.000	1.41
1,2,3,4,7,8,9-HpCDF	0.001	0.20	0.000	0.28	0.000	0.00
OCDF	0.000	0.00	0.000	0.04	0.000	0.00
PCDFs	0.506	82.26%	0.076	78.41%	0.019	79.58%
PCDDs+PCDFs	0.615	100%	0.097	100%	0.024	100%

Note: The figures in shaded areas represent the compositional percentile of each 2,3,7,8 congener to total TEQs.

78°C), respectively. Also, the off-gas per ton of product was averaged about 25,863 Sm³/ton ranging from 9,551~58,365 Sm³/ton, and the major APCD was all bag filter. As in the case of steel-melting facilities, most of electric alloy-melting facilities emitted the lots of off-gases up to about 1.3 millions standard cubic meters per hour, and also off-gases contained 20.1% of oxygen and 5,011 ppm of carbon monoxide on average. This is the same reason caused by the sequence-batch operation as in steel-melting furnace.

PCDDs/PCDFs emissions were in the range of 0.001 to 0.052 ng-TEQ/Sm³ (average 0.024 ng-TEQ/Sm³, s=0.135) from six measurements. Like emission patterns of sintering and steel-melting furnaces, PCDFs from alloy-melting furnaces showed approximately four times higher concentrations than PCDDs, showing that the ratio of PCDDs to PCDFs was 20:80 on average. The most abundant 2,3,7,8-congener was also 2,3,4,7,8-PeCDF followed by 1,2,3,7,8-PeCDD, and their TEQ values constituted 47.2% and 8.5% of total TEQs,

respectively. 2,3,7,8-TCDD was detected at a level of 7.8% of total TEQs.

4. Conclusions

Four iron-ore sintering furnaces, thirteen steel-melting furnaces, and three alloy-melting furnaces were selected from eighteen plants in iron-and-steel foundries. PCDDs/PCDFs samplings were achieved two times at the stacks of each furnace in order to investigate the emission characteristics or patterns of PCDDs/PCDFs in the sintering and melting processes. Results could be summarized as follows;

1. From a total of 36 measurements in iron-and-steel foundries, PCDDs/PCDFs emission levels were in the range of 0.024 to 0.615 ng-TEQ/Sm³ on average. PCDFs showed an approximately four times higher concentrations than PCDDs, revealing that the ratio of PCDDs to PCDFs averaged approximately 20:80. The main contributor to total TEQs was 2,3,4,7,8-PeCDF,

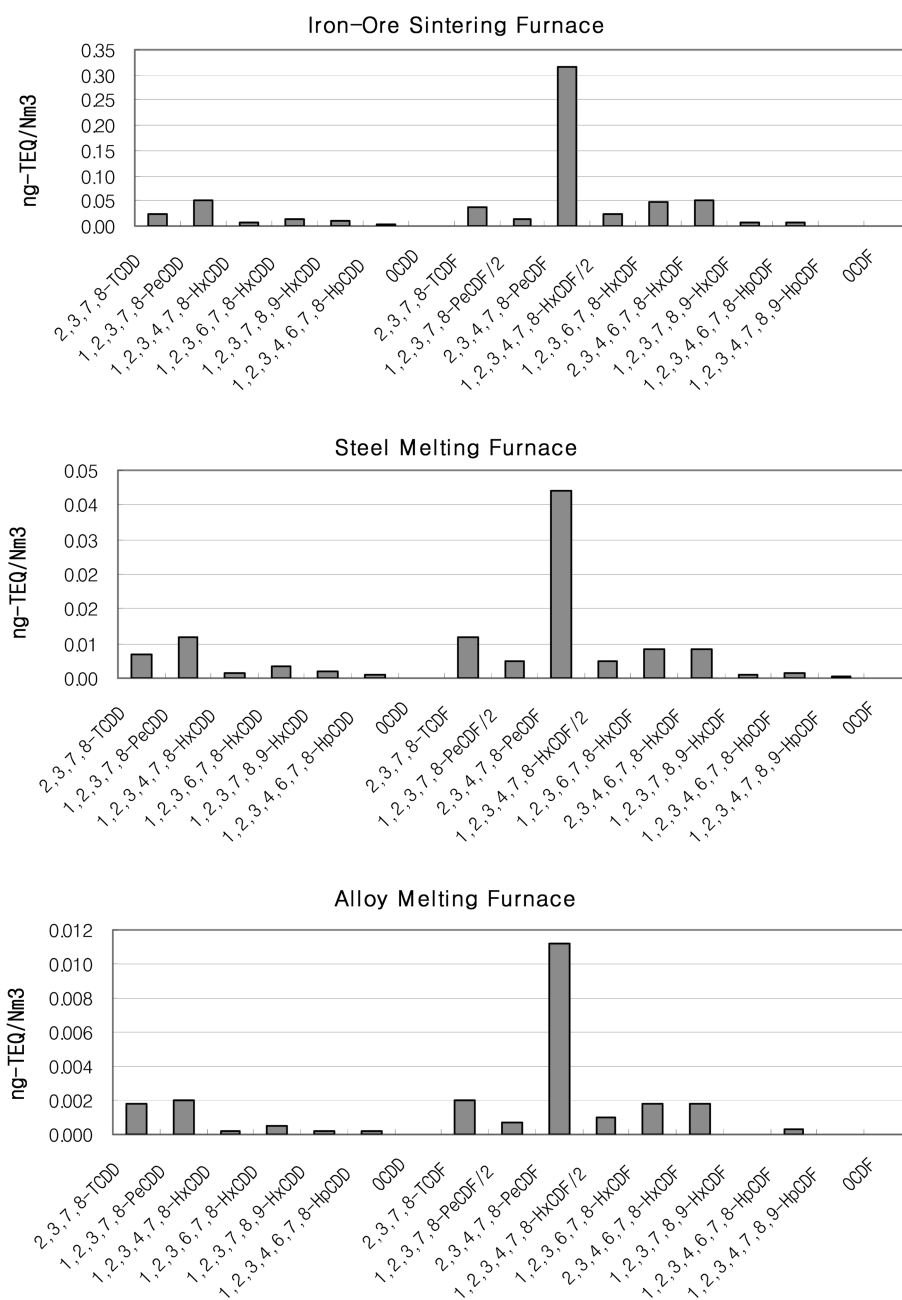


Fig. 2. Congener profiles in sintering, steel-melting and alloy-melting furnaces.

and its TEQ values were in the range of 44% to 52% of total TEQs.

2. The major problem of sintering furnace is that it emits one million or more standard cubic meters of off-gases per hour, and the off-gas released into atmosphere contains approximately 1.6% carbon monoxide which

can not be controlled by electrostatic precipitator.

3. The sequence-batch operation of electric furnace results in lots of off-gas generation and a very high concentration of oxygen in the flue gas stream, showing an average of 1.5 millions or more standard cubic meters of off-gas, 20.5% of oxygen and 410 ppm of

carbon monoxide in the electric steel-melting furnaces, and 1.3 millions or more standard cubic meters of off-gas, 20.5% of oxygen and 410 ppm of carbon monoxide in the electric alloy-melting furnaces, respectively.

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