EFFECT OF FURNACE TYPE AND SCRAP QUALITY ON DIOXIN EMISSION FROM ALUMINUM REFINEMENT IN JAPAN

Takeuchi M¹, Nakayama T²

¹Energy Technology Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), 16-1 Onogawa, Tsukuba, 305-8569 Japan, ²Nonferrous Metals Division, Manufacturing Industries Bureau, Ministry of Economy, Trade and Industry, 1-3-1 Kasumigaseki, Chiyoda-ku, Tokyo, 100-8901 Japan

Introduction

In the recycling process of aluminum scrap, dioxins (PCDD/Fs and DL-PCB) are certainly formed around the melting process of the scrap and other raw material. The reason of dioxins formation has not been understood exactly yet, however, the carbon skeleton of dioxins is probably produced with the breakdown of soot or PAHs that produced in the combustion of burner fuel and organic matter in the scrap¹). Chlorine contained in the scrap and injected in the refining process might also act as a dominant roll on the formation²). We suggested in the previous report³ that there is the strong relationship between chlorine contents in the scrap and the dioxins concentration in the flue gas of the melting furnace. However, these results were obtained using a single experimental furnace, and we still don't know that the same results could obtain or not using various furnaces in operation. Usually, chlorine content varies with type of the scrap, and low quality scrap contains many organic matters like plastics and paints. Moreover, the type of furnace that melts aluminum scrap is selected according to the form and quality of scrap. Then, this study has been conducted to clarify the effects of furnace type and scrap quality on the dioxins emission level from the melting furnaces.

Materials and Methods

The dioxin law (the law concerning special measures against dioxins, 1999) of Japan specifies emission standards for aluminum refinement (precisely, the standards applies all facilities for manufacturing aluminum base alloy). That is, 1 ng-TEQ/m³ is applied to furnaces newly constructed after December 2002, while 5 ng-TEQ/m³ is applied to existing furnaces. Moreover, the measurement of dioxins in the designated facilities, including the aluminum scrap melting furnaces, must be performed once a year in accordance with the dioxin law. The measured values of dioxins are open disclosed to the public. The measurement is based on JIS (Japanese Industrial Standard) that requires the employment of HR-GC/MS and WHO-TEF (1998).

In this study the data are obtained from the questionnaire inquiries conducted by the Ministry of Economy, Trade and Industry in July and September 2005. Based on concentration data of dioxins officially announced mandatory in 2004 in accordance with the law, the inquiries were made on the types of furnaces, materials used, equipped countermeasure facilities for dioxins reduction, and the amount of production at each factory site. The concentration of dioxins emission levels and types of furnace purpose; melting, drying and delacquering, were surveyed in July, and the detailed inquiry of the melting furnaces; types, materials used and countermeasures in relation to dioxins, were conducted in September. Table 1 shows the detail of response situation of the inquiries. Though the inquiry contained not only melting furnaces but also drying furnaces and delacquering furnaces, this report treats only the results of melting furnaces.

Type of furnaces	Number of companies (factory sites) investigated	Number of companies (factory sites) that responded in July 2005	Response rate (%) in July 2005	Number of companies (factory sites) that responded in September 2005	Response rate (%) in September 2005
Melting furnace		73 (88)		59(70)	79.5
Rotary type drying furnace	98 (114)	20 (24)	74.5 (77)	17(21)	87.5
Used can delacquering (decoating) furnace		10 (10)		10(10)	100

Fable 1	The	extent	of	inquiry	and	response	rates.
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Figure 1 Dioxins concentration distribution in emission of melting furnaces.

Results and Discussion

1) Dioxins emission levels of the melting furnaces.

All dioxins concentration data of the melting furnaces are classified to the levels of concentration in figure 1. About 80% of the furnaces have a value under 0.5 ng-TEQ/m³, and almost 41% achieves under 0.1 ng-TEQ/m³. There are still many furnaces that emitted over 0.5 ng-TEQ/m³ and even 1 ng-TEQ/m³. Fortunately, since newly build furnaces are very few, the emission values did not exceed the current regulation. However, we must consider the reason that still exist the higher level furnaces, although aluminum refining companies has been continuously tried to decrease the dioxins emission level with many efforts, such as addition of dioxin control facilities to the furnaces.

2) Types of melting furnaces

Table 2 shows number of furnaces operating and amounts of production on a furnace structure basis in Japan. About 90% of aluminum alloy is produced from closed-type reverberatory furnaces and those with open wells. Obviously, over 70% of alloy is produced from the furnaces with open wells. Then analysis of operation situations of the furnaces with open well might be useful in Japanese case.

	Number of each		Amount of production	
	furnace	%	(ton/y) in 2004	%
Reverberatory furnace with open	101	46.3	1,050,054	71.5
Closed-type reverberatory furnac	37	17	249,631	17
Holding furnace	28	12.8		
Rotary furnace	40	18.4	127,662	8.7
Pot furnace	5	2.3	3,400	0.2
Low frequency induction furnace	3	1.4	12,250	0.8
Others(melting holding furnace)	4	1.8	25,240	1.7
Total	218	100	1,468,237	100

Table 2 Furnace types used and production rats in Japan.

Holding furnaces are not used independently.

Table 3 shows the dioxins emission level from each melting furnace type separately. The results indicate that emission level of the closed type furnaces and the rotary furnaces distributed mainly lower level range. On the other hand, many furnaces with open well seem to have rather higher emission level such as over 0.5ng-TEQ/m³. The structure of the furnace with the open well consists of two sections; a smelting chamber heated by a heavy oil burner connecting through a molten aluminum path to the open well where aluminum scrap are supplied. If the scrap contains a large number of small and thin fragments, direct supply to the heated chamber would cause in excess oxidized loss because they will be exposed directly to the open well. It is said that the selection of furnace type is strongly related to the kind of expected scrap. Therefore, the furnace with open well has a tendency to accept wide variety of scrap especially low quality one which contains many small fragments or unselected scrap such as construction waste.

3) Effect of the scrap quality

Table 4 shows the detail of the supplied raw materials to the melting furnaces. Municipal scrap contains a many impurities such as plastics including polyvinylchloride and its degree of contamination depends on the collection condition. Clearly, the furnaces with open well have accepted wider variety of scrap including municipal scrap than other furnace type. Table 4 also shows the average dioxins concentration of furnaces using municipal scrap is obviously higher than that without municipal scrap. The closed type furnaces and the rotary type furnaces emitted lower level dioxins than furnaces with open well, however, their emission concentrations also increase when the municipal scrap were supplied. Then, we might say it is confirmed that the difference in dioxins concentration from melting furnaces mainly depends on the quality of the aluminum scrap, not on the furnace type.

		Reverberatory	Closed type furnace	Rotary furnace
		furnace with open		
furnaces reported*		73	18	23
Range of dioxins	0.1>x	21	10	14
emission : ng-TEQ/m ³	0.2>x>=0.1	12	2	7
- · ·	0.3>x>=0.2	11	1	0
	0.4>x>=0.3	3	0	1
	0.5>x>=0.4	4	3	0
	0.6>x>=0.5	6	0	0
	0.7>x>=0.6	6	1	0
	0.8>x>=0.7	2	0	0
	0.9>x>=0.8	2	0	0
	1.0>x>=0.9	0	0	0
	x>=1.0	6	1	1
Average concentration	Average, (o), upper	0.38, (o=0.47),	0.23, (σ=0.31),	0.17, (o=0.45), 0.36/0
of dioxins in ng-TEQ/m ³	limit/lower limit of 95%	0.49/0.28	0.37/0.08	
	confidence interval			
Operating conditions of	Melting	35	6	7
measurement	Melting and Refinement	25	10	9
	Melting, Holding and Casting	6	0	0
	Melting, Refinement, Holding and Casting	4	0	0
	Refinement, Holding and Casting	1	1	2
	Melting and Dross treatment	2	1 (Suspended)*	5
Types of production		Secondary Al alloy ingot for casting and die casting (70%).	Mainly secondary Al alloy ingots (75%). Molten metal supply is in 4 furnaces (22.2%).	Mainly base metal production (including molten metal supply) (11 cases, 47.8%). Secondary Al alloy ingots (10cases, 43.5%).

Table 3 Detail of dioxins emission levels and operatiing condition of furnaces at measurement time.

* Only dross treatment equipment is being operated.

		Reverberatory furnace with open well	Closed-type Reverberatory furnace	Rotary furnace
Total number of furnaces reported*		73	18	23
Types of supplied raw	Municipal scrap	64	10	17
materials at	Factory scrap	36	7	9
measurement time	Scrap from pre-treatment	27	4	1
	Primary aluminum	23	4	2
	Base metal	53	12**	1
	Molten metal	7	1	-
	Dross	-	-	5
Average in ng-TEQ/m ³ :		64	10	16***
The case of municipal scrap used	Average, (ơ), upper limit/lower limit of 95% confidence interval	$0.41, (\sigma = 0.49), 0.53 \\ \checkmark 0.30$	$0.33, (\sigma = 0.37), 0.56 \\ \checkmark 0.11$	0.09, ($\sigma = 0.09$), 0.13 / 0.05
Average in ng-TEQ/m ³ :		9	8	6***
The case of municipal scrap is not used	Average, (o), upper limit/lower limit of 95% confidence interval	0.15, (σ =0.15), 0.25 \checkmark 0.05	0.09, (σ =0.16), 0.20 \checkmark 0.0	0.05, ($\sigma = 0.09$), 0.12 $\checkmark 0.0$
Countermeasure	Pre-treatment of raw materials	22	4	4
facilities used	Cyclone dust collection	52	9	12
	Bag filter dust collection	69	16	20
	Wet-type dust collection	2	1	3
	Catalytic filter	1	3	-
	Slaked lime injection	24	-	5
	Activated carbon injection	4	-	-
	Use low-discharge flux.	-	1	-
	Both cyclone and bag filter used	51	9	12
Notes		For 6 examples in which the dioxins exceed 1ng-TEQ/m ³ , municipal scrap is used.	Usually, scarp quality is better than furnaces with open well.	This type is usually used for recovering metal from dross or for base metal production.

Table 4 Detail of supplied raw material in conection with dioxin emision level.

* Only furnaces with single dust-collecting unit. Furnaces with common dust collectors with other furnaces are omitted.

** Only base metal is used in 2 furnaces.

*** Statistical process was performed excluding 1 sample that municipal scrap (wrecked scrap with iron) used.

Already almost all melting furnaces in Japan are equipped and applied various countermeasure facilities and methods. However, there is no apparent deference between applied countermeasure on the dioxins emission levels. We still continue the data collection and will try to analyze it in next step.

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References

- 1 Iino F, Imagawa T, Takeuchi T. Sadakata M *Environ Sci Technol* 1999;33:1038
- 2 Hatanaka T, Imagawa T, Takeuchi M. Environ Sci Technol 2000;34:3920
- 3 Takeuchi M, Oya M, Matsunaga S, Ozono T. Organohalogen Comp 2002;56:213