



# **RECYCLING OF WASTE PLASTICS DISPOSED OF IN LANDFILLS:** THE EFFECT OF WASHING TREATMENT

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### **ABSTRACT**

In recent years, environmental pollution in the vicinity of final disposal sites for inert waste has resulted in problems such as deterioration of water quality and increases in leachates and odours. Accordingly, the number of sites that no longer accept these wastes has risen. One of the main causes of these problems is represented by organic matter adhering to waste plastics, accounting for the majority of waste deposited at final disposal sites for inert waste. Furthermore, the amount of waste plastics undergoing thermal recycling has increased owing to recent energy demands. This study examines the recycling of waste plastics into solid fuel, known as refuse paper, and plastic fuel (RPF), by focusing on the washing method. The first washing effect at liquid to solid ratio (L/S) = 1 was confirmed to be effective in the rough removal of adherent organic matter and was also found to be suitable for pretreatment for the second washing for thermal recycling. As compared with the first washing, although effectiveness of the second washing with regard to chemical oxygen demand (COD) was not observed, the effectiveness of the second washing versus total nitrogen (T-N) at low speed and L/S = 1 was comparable to that of the first washing at L/S = 10. These results indicate that a combination of first washing at L/S = 1 and second washing at low speed and L/S = 5 constituted an effective pretreatment for refuse paper and plastic fuel (RPF) production. Furthermore, a mixed antichlor was produced by combining a commercial antichlor and waste having a dechlorination effect as countermeasures against hydrogen chloride gas emitted when RPF derived from the burning of waste plastics. We tested its performance in reducing Cl gas generation by mixing it into the RPF. As the antichlor was added to RPF for low Cl gas emission, 36% of the CaO-oyster shells added as antichlor against CI content was found to be effective in reducing CI gas emission. Thus, it was concluded that using an appropriate percentage of antichlor, the combination of first and second washing processes (under some conditions) was effective in the pretreatment of waste plastic destined to be recycled as RPF.

### **1. INTRODUCTION**

In recent years, environmental pollution in the vicinity of final disposal sites for inert waste has resulted in problems such as deterioration of water quality and increases in leachates and odours. Accordingly, the number of sites that no longer accept these wastes has risen. Although organic matter adhering to waste plastics, accounting for the majority of waste deposited at final disposal sites for inert wastes, is considered to be a cause of these problems, the amount of incoming wastes with these characteristics is controlled simply by visual inspection, as required by law (Act on the Technology Standards for final disposal of municipal solid waste and industrial solid waste, (article 2.2). Following implementation of the revised Waste Management Act, enforced on June 17, 1998, the installation of new facilities including final disposal sites for inert waste has advanced smoothly due to the prescribing of structural reinforcement and a complicated application procedure. Therefore, the number of newly installed final disposal sites for inert wastes has decreased since 1999 (Ministry of the Environment, 1998-2012). Furthermore, the





amount of waste plastics recycled increased by 2.3 × 105 tons from FY2011 to FY2013. As a result, the amount finally disposed of decreased by about 3.1 × 105 tons from 10.5 × 105 tons in FY2011 to 7.4 × 105 tons in FY2013. Of the many recycling categories, only thermal recycling, including refuse-derived fuel, raw material for cement production, and fuel used in cement kilns, increased from FY2011 to FY2013, at 5.3 × 105 tons (Plastic Waste Management Institute, 2012, 2015). The use of waste plastics as an alternative fuel has therefore increased in the light of recent energy demands. Moreover, prior to enforcement of the Law for the Promotion of Sorted Collection and Recycling of Containers and Packaging in April 2000, good-quality waste plastics, which should be recycled, were deposited in final disposal sites for inert wastes. These good quality waste plastics are considered to be suitable candidates for thermal recycling or material recycling, deemed to be important resource repositories for resource-poor Japan. However, currently enforced laws and the manifest system make it impossible to exhume inert wastes and to recover waste plastics for recycling from the final disposal sites. Therefore, an amendment to these laws to exhume inert wastes and to recover waste plastics for recycling from the final disposal sites is mandatory.

EU laws establish that recycling of all waste plastics by the year 2030, with residues being landfilled directly at the final disposal site. The current state of the art for recycling of waste plastics in Asia is still significantly lacking.

This paper examines the recycling of waste plastics deposited in final disposal sites for inert wastes prior to April 2000 and waste plastics disposed of in landfills. Waste plastics that are not recycled but disposed of in landfills may result in foul odours during transportation and storage due to the presence of adherent organic matter. Similar odours also occur at final disposal sites for inert wastes. Focusing on an effective washing method as a pretreatment technique for incineration residue (Tameda et al., 2007), we developed a technique for use in the efficient recycling of waste plastics. Our washing method includes two steps. The first washing relates to on-site agitation in water tanks to remove adherent organic matter and resolve the issue of odours during transportation and storage at final disposal sites for inert wastes. The second washing is performed by means of high-speed rotation to thoroughly remove adherent organic matter and enable the use of waste plastics for thermal recycling.

In the recycling of waste plastics, a solid fuel known as refuse paper and plastic fuel (RPF) was used, thus facilitating its subsequent use as an alternative fuel. It should be noted that hydrogen chloride (HCI) gas is emitted when RPF is burned. The content of Cl in RPF, a cause of gas emission, is regulated by Japanese Industrial Standard (JIS) Z7311:2010) as a mass fraction of all Cl content. However, some types of RPF do not meet this standard, and countermeasures against Cl gas are required. A mixed antichlor was therefore produced by combining a commercial antichlor and waste having a dechlorination effect, and its performance in reducing Cl gas generation was tested by mixing it into the RPF.

### 2. MATERIALS AND METHODS

# 2.1 Experiment on the effect of first washing to roughly remove adherent organic matter

We conducted an experiment on the effects of the first washing to roughly remove adherent organic matter.

### 2.1.1 Overview of the experiment

For the first washing experiment, 40-foot containers were modified to allow agitation from above using a heavy machine (Figure 1). Two water tanks were installed, including the first and second washing tanks. 15 m<sup>3</sup> of water was poured into the first and second tanks, respectively, for a total of 30 m<sup>3</sup>. The liquid-solid ratio (L/S) was determined as the volume of water against the volume of waste to be washed in a 1:1 ratio. For this experiment, waste containing waste plastics that would be deposited in final disposal sites for inert waste was used; 10 batches of 3 m<sup>3</sup> waste were continuously washed. The time required for washing once (at the first washing tank + the second washing tank) corresponded to approx. 30 minutes and it did not affect the subsequent capitalization process. To assess washing efficiency, waste plastics were extracted from the waste for analysis. We analysed the compositions of the waste to be washed and performed chemical oxygen demand (COD) and total nitrogen (T-N) on the waste plastics before and after washing (Table 1).

### 2.1.2 Results and Discussion

Ten cases in which inappropriate matter for disposal was contained in waste were selected by visual inspection and the compositions analysed on the basis of weight percentage; the results are shown in Table 2. Overall, waste plastics accounted for 63% on average, and inappropriate matter for disposal other than the five types of stable wastes such as waste plastics, rubber scrap, metal scrap, demolition wastes and waste glass and ceramics accounted for 14% on average. The details of Case 9, used for the first washing experiment, are shown in Figure 2. Waste plastics constituted the highest percentage at 35%; inappropriate matter was second at 31%; and residue of 75 mm or smaller were third, at 24%. Polyvinyl chloride (PVC), an undesirable compound in thermal recycling, was as low as 2%. Organic pollutants such as wood, paper, and food residues contained in the inappropriate matter, likewise undesirable for landfills, corresponded to 11.8%.

Figure 3 shows the results of a leachate test for the COD of waste plastics samples collected before and after the first washing experiment. The value obtained before first washing was 170 mg/L. After washing, values of 27 mg/L (L/S = 10; removal efficiency = 83%), 29 mg/L (L/S = 3; removal efficiency = 83%), 35 mg/L (L/S = 1.5; removal efficiency = 79%), and 39 mg/L (L/S = 1; removal efficiency = 77%) were obtained. These results indicate that although COD removal efficiency decreased as L/S decreased, adequate washing efficiency was obtained even at L/S = 1.

Figure 4 shows the results of a leachate test for T-N of waste plastics samples collected before and after the first washing experiment. The value obtained prior to test-ing was 16 mg/L. Subsequently, values of 2.1 mg/L (L/S



FIGURE 1: Outline of first washing procedure.

= 10; removal efficiency = 87%), 4 mg/L (L/S = 3; removal efficiency = 75%), 3.5 mg/L (L/S = 1.5; removal efficiency = 78%), and 3.6 mg/L (L/S = 1; removal efficiency = 78%) were obtained. These results indicate that although T-N removal efficiency decreased as L/S decreased, adequate washing efficiency was obtained even at L/S = 1. After washing ten times, the residual water COD-Mn was 120 mg/L and T-N was 19mg/L. Depending on its quality the residual water was discharged or recycled.





# 2.2 Experiment on the effect of second washing to produce RPF

The recycling of waste plastics into RPF was investigated. To further remove adherent organic matter on waste plastics roughly washed by first washing, we adopted a second washing process as pretreatment for RPF production and conducted an experiment on the effects of the second washing process.

TABLE	1:	Plan	of	first	washing	experiment.
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Sampling interval of first washing tank	After 1st, 3rd, 5th, 7th and 10th washing		
Sampling interval of second washing tank	After 1st, 3rd, 5th, 7th and 10th washing		
	COD-Mn T-N the standard methods = Ministry of the Environment Notification No. 46		
Leachate test items for waste plastics before and after washing	The amount of the sample = solid (1): liquid (10)		
	The number of replica = 3 replicas		
Waste to be washed	Composition analysis		

**TABLE 2:** Results of composition analysis of waste to be washed.

	Waste plastics		Dem-	Ceramics		Inappro-	Res	idues	Total	Measured weight	
	Soft	Non soft	waste	waste	Metals	matter	(2 mm~ 75 mm)	(2 mm or smaller)	•••A	•••B	A-B
Case 1	101.4 25.1%	249.6 61.8%	0.1 0.0%	-	2.6 0.6%	16.0 4.0%	30.8 7.6%	3.2 0.8%	403.7 100.0%	440	-36.3 91.75%
Case 2	47.2 11.2%	304.0 72.4%	-	-	4.0 1.0%	46.4 11.0%	17.6 4.2%	0.8 0.2%	420.0 100.0%	420	0.0 100.00%
Case 3	64.0 9.5%	379.2 56.3%	9.6 1.4%	-	32.0 4.7%	77.2 11.5%	105.6 15.7%	6.4 0.9%	674.0 100.0%	960	-286.0 70.21%
Case 4	68.0 23.9%	66.4 23.3%	20.8 7.3%	-	12.0 4.2%	36.8 12.9%	47.2 16.6%	33.6 11.8%	284.8 100.0%	290	-5.2 98.21%
Case 5	26.4 15.0%	62.0 35.2%	0.8 0.5%	-	1.2 0.7%	44.4 25.2%	36.8 20.9%	4.4 2.5%	176.0 100.0%	190	-14.0 92.63%
Case 6	36.8 7.2%	218.4 42.9%	19.2 3.8%	15.2 3.0%	23.2 4.6%	124.0 24.3%	64.8 12.7%	8.0 1.6%	509.6 100.0%	620	-110.4 82.19%
Case 7	222.4 20.7%	700.8 65.4%	12.8 1.2%	-	30.4 2.8%	25.6 2.4%	75.2 7.0%	4.8 0.4%	1,072.0 100.0%	1290	-218.0 83.10%
Case 8	101.6 10.9%	357.6 38.5%	106.4 11.5%	35.2 3.8%	32.0 3.4%	52.4 5.6%	226.4 24.4%	17.6 1.9%	929.2 100.0%	830	99.2 111.95%
Case 9	59.2 6.7%	272.8 31.0%	24.0 2.7%	16.0 1.8%	26.4 3.0%	273.6 31.1%	168.0 19.1%	40.8 4.6%	880.8 100.0%	1030	-149.2 85.51%
Case 10	100.8 17.6%	326.4 56.9%	4.0 0.7%	-	11.2 2.0%	50.4 8.8%	79.2 13.8%	1.6 0.3%	573.6 100.0%	610	-36.4 94.03%
											Unit: Kg



FIGURE 3: Results of chemical oxygen demand (COD) - leachate test before and after washing.

### 2.2.1 Overview of the experiment

For the second washing experiment, a high-speed rotary washer (Photo 1) was used and performed leachate tests of waste plastics collected before and after the second washing to check the effectiveness of the second washing process under several washing conditions. The washing conditions used in this experiment are shown in Table 3. COD value before the first washing was 170 mg/L. After washing, values of 39 mg/L (L/S = 1; removal efficiency = 77%) were obtained. Thus, to check the effects of removal in second washing process, organic matter adhering to waste plastics that were inappropriate for disposal in landfills and were unwashed before the second washing experiment were used as samples.

### 2.2.2 Results and Discussion

Table 4 shows the removal efficiency of biochemical oxygen demand (BOD), COD, and T-N for waste plastics after the second washing. The influence of L/S on BOD removal efficiency was inconsistent. Although BOD removal efficiency tended to be higher as L/S became high at low speeds, it increased in the order of L/S = 3, 1, and 5 at me-



PHOTO 1: Rotary washer for second washing.



FIGURE 4: Results of total nitrogen (T-N) - leachate test before and after washing.

dium speed and L/S = 5, 3, and 1 at high speed. The influence of number of revolutions (speed) on BOD removal efficiency was such that although BOD removal efficiency was highest at low speed at L/S = 3 and 5, it was also high at medium speed at L/S = 1. The influence of L/S on COD removal efficiency was likewise inconsistent. Although COD removal efficiency tended to be higher as L/S became high at low speed, it increased in the order of L/S = 3, 1, and 5 at medium speed. At high speed, it was highest at L/S = 1, whereas COD removal efficiency when L/S = 3 was equal to that obtained at L/S = 5. The influence of the number of revolutions (speed) on COD removal efficiency was such that COD removal efficiency was highest at low speed. The influence of L/S on T-N removal efficiency was observed at low speed.

As an overall tendency observed in the results described above, washing efficiency achieved highest values when the number of revolutions was low and L/S = 5. As compared with the first washing, although the effectiveness of second washing on COD was not observed, the influence produced on T-N at low speed and L/S = 1 was comparable to the effectiveness of first washing at L/S = 10. These results indicate that a combination of first washing with L/S = 1 and second washing at low speed and L/S = 5 represents an effective pretreatment for RPF production.

### 2.3 Experiment on antichlors

An experiment was conducted to examine the antichlors of reducing HCl gas generated in burning of RPF produced from waste plastics subjected to the second washing. A standard concentration was established for HCl gas present in gas emitted from RPF of JIS grade A, in which Cl content was 0.3% or less. The final goal was to meet the standard while using an RPF Cl content of 0.5%. This value corresponds to an RPF of JIS grade B, in which Cl content exceeds 0.3% but lower than 0.6%. To observe the dechlorination effect, we used PVC, which has a high conversion ratio to HCl, rather than RPF (Kawamoto et al., 2011). TABLE 3: Conditions of second washing experiment.

Number of revolutions of high-speed rotary washer	700 rpm (low speed), 880 rpm (medium speed), 1250 rpm (high speed)
Particle size of waste plastics to be washed	40 mm
Liquid - solid ratio	1 time, 3 times, 5 times
Matter to be analyzed	Waste plastics to be washed regarded as inappropriate for final disposal at such sites for inert waste. Waste plastics before and after washing; wastewater after washing.
Items analyzed (waste plastics before and after washing)	pH, EC, Cl, COD, BOD, T-N, T-C, TOC, IC
Items analyzed (wastewater after washing)	pH, COD, BOD, T-N, T-S, S <sup>2-</sup> , SS, T-P

TABLE 4: Removal efficiency of biological oxygen demand (BOD), chemical oxygen demand (COD) and total nitrogen (T-N) by second washing.

Liquic	d-solid ratio	1 time	3 times	5 times	1 time	3 times	5 times	1 time	3 times	5 times
Number of revolutions			Low speed			Medium spee	d		High speed	
	Before washing (mg/L)					56				
BOD	After washing (mg/L)	13	8.8	6.6	9.2	13	7.9	14	13	17
	Removal efficiency (%)	77%	84%	88%	84%	77%	86%	75%	77%	70%
	Before washing (mg/L)					110				
COD	After washing (mg/L)	37	25	23	33	47	26	49	56	56
	Removal efficiency (%)	66%	77%	79%	70%	57%	76%	55%	49%	49%
	Before washing (mg/L)		•	•		64	•			
T-N	After washing (mg/L)	2.3	2.1	2.3	4.8	8.4	3.1	8.2	8.9	8.3
	Removal efficiency (%)	96%	97%	96%	93%	87%	95%	87%	86%	87%

### 2.3.1 Overview of the experiment

Nine agents and seven types of waste (Table 5) were selected as antichlors, and their dechlorination effects when used alone were investigated as single-antichlor experiments. Several agents and wastes featuring high dechlorination efficiency were selected to create mixed antichlors by combining an agent and a type of waste, and their dechlorination effects were investigated to determine the best antichlor to use in mixed-antichlor experiments.

The experiments were performed using the equipment illustrated in Figure 5. PVC and an antichlor in a porcelain dish were placed in an electric furnace. Air was introduced into the furnace by an air pump, and the emitted HCl gas was trapped in a flask. HCl gas concentration was measured by redox titration in an HCl-trapping experiment. The burning conditions of the experiment are shown in Table 6. Other properties of the emission gas were also investigated by a simplified method in an experiment conducted to assess emission gas properties.

### 2.3.2 Results and Discussion

2.3.2.1 HCI-trapping experiment. To develop new antichlors,

the amount of HCl gas emitted and removal efficiency of HCl gas were obtained by using single antichlors, as listed in Table 5; the results are shown in Figures 6 and 7 and in Table 7. Agents meeting the standard included CaO, NaH- $CO_3$ , and ZnO. CaO displayed the highest removal efficiency at about 88%. Although none of the waste types met the standard, calcined lime cake displayed the highest removal



FIGURE 5: Schematic illustration of combustion experiment equipment.

TABLE 6: Conditions o	HCI-gas-trapping	experiment
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Temperature	800 °C
Combustion retention time	10 min
Supplied air flow	700mL/min
Air flow time	10 min
Analysis method	Redox titration

#### TABLE 5: Selected antichlors.

Agents	Calcium carbonate, magnesium oxide, mag- nesium hydroxide, calcium oxide, calcium phosphate, calcium hydrogen phosphate, sodium hydrogen carbonate, zinc oxide, magnesium hydrogen carbonate
Waste	Lime, oyster shells, zeolite, scallop shells, neutral solidifying agent, lime cake, calcined lime cake



(agents).

efficiency of about 18%, followed by lime cake 13%, oyster shells 9%, and scallop shells 7%.

Subsequently, the amount of antichlor to be added was examined to enhance removal efficiency for HCl gas. However, it proved difficult to increase the quantity of the agent due to economic reasons. Thus, only the four waste-derived antichlors featuring the highest removal efficiency, as previously described, were examined, and the amounts were set at 10%, 20%, 30%, and 40% of Cl content. It should be noted that the amount of calcinated lime cake was only 40% owing to amount limitations. The results, shown in



FIGURE 7: Amount of HCl gas emitted with single antichlors (waste).

Figure 8 and Table 8, indicated that oyster shells (40%) displayed the highest removal efficiency at approx. 60%.

On the basis of these results, we selected CaO from the agents and oyster shells from the waste for the mixed antichlor, and set the agent–waste ratios at 0.5%-3.6%, 1%-1.8%, and 1%-36% considering the synergistic effect. The results of the combustion experiment of mixed antichlors are shown in Figure 9 and Table 9. As a result, only the antichlor of CaO–oyster shells with 1% and 36% added against the Cl content, respectively, met the standard, displaying a removal efficiency of approx. 71%.

TABLE 7: Removal efficiency of HCl gas with single antichlors (agents or waste).

Single antichlor (agents)	Removal efficiency	Single antichlor (agents)	Removal efficiency	
Calcium carbonate	31.50%	Magnesium hydrogen carbonate	49.05%	
Magnesium oxide	5.49%	Lime	5.45%	
Magnesium hydroxide	45.11%	Oyster shells	8.59%	
Calcium oxide	87.83%	Zeolite	-0.65%	
Calcium phosphate	23.15%	Scallop shells	7.48%	
Calcium hydrogen phosphate	24.70%	Neutral solidifying agent	2.49%	
Sodium hydrogen carbonate	64.44%	Lime cake	13.20%	
Zinc oxide	81.74%	Calcined lime cake	17.82%	



FIGURE 8: Amount of HCl gas emitted by addition ratio of single antichlors (waste).



FIGURE 9: Amount of HCl gas emitted with mixed antichlors.

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**TABLE 8:** Removal efficiency of HCl gas by addition of single antichlors (waste).

Single antichlor (waste)	Removal efficiency
Scallop shells 10	16.90%
Scallop shells 20	19.67%
Scallop shells 30	24.28%
Scallop shells 40	30.38%
Oyster shells 10	17.82%
Oyster shells 20	29.46%
Oyster shells 30	51.43%
Oyster shells 40	59.74%
Lime cake 10	17.08%
Lime cake 20	22.44%
Lime cake 30	40.17%
Lime cake 40	52.17%
Calcined lime cake 40	29.82%

TABLE 9: Removal efficiency of HCL gas with mixed antichlors.

Mixed antichlor	Removal efficiency
Calcium oxide 0.5% + Oyster shells 3.6%	36.29%
Calcium oxide 1% + Oyster shells 1.8%	35.34%
Calcium oxide 1% + Oyster shells 36%	71.38%

2.3.2.2 Experiment on emission gas properties. Using the selected antichlor, the properties of the emission gas were analysed by means of a simplified method; the results are shown in Table 10. Although HCl gas concentration of a blank sample with a Cl content of 0.5% was 3,300 mg/m<sup>3</sup>N, the concentration detected in a sample with the mixed antichlor was as low as 260 mg/m<sup>3</sup>N, with a calculated removal efficiency of 92%. Indeed, the antichlor featuring a combination of 1% CaO to 36% oyster shells added against Cl content was found to act as an effective mixed antichlor,

and the amount of antichlor used to treat CI gas in emission gas was thus reduced.

# **3. CONCLUSIONS**

The results of this study are summarized in the following points:

- The effect of the first washing at L/S = 1 was confirmed, as was the effect of first washing to roughly remove adherent organic matter. The first washing was also found to be suitable for use as pretreatment prior to second washing for thermal recycling.
- As an overall tendency observed in waste plastics after the second washing, washing efficiency was found to be highest when the number of revolutions was low (700 rpm) and L/S was 5. With regard to the influence of L/S at each number of revolutions on removal efficiency, a higher L/S related to a higher removal efficiency of organic pollutants.
- 3. As compared with first washing, although the effectiveness of the second washing against COD was not observed, the effectiveness of second washing against T-N at low speed and L/S = 1 was comparable to that obtained with the first washing at L/S = 10. These results indicate that a combination of first washing with L/S = 1 and second washing at low speed and L/S = 5 constitutes an effective pretreatment for RPF production. Moreover, to lengthen the second contact time of washing water and waste plastics in the high-speed rotary washer, an increased cleansing efficiency may be expected due to a "gradual change in the rotational speed at a low-speed side" and by "soaking washing".
- 4. As an antichlor to be added to RPF with low Cl gas emission, the addition of 1% CaO and 36% oyster shells against Cl content was found to be effective, and the amount of antichlor used to treat Cl gas in the emission gas was thus reduced.

A	IA	11	Sample name		
Analysis	items	Units —	Blank sample	Sample with mixed antichlor	
	Average temperature of gas in furnace	Measured value	°C	808	810
Emission	Moisture content	Measured value	%	9.3	10.5
	D	Measured value	g/m³ <sub>N</sub>	1.7	1.8
	Dust concentration	O <sub>2</sub> - reduced value	g/m³ <sub>N</sub> •12%	5.5	5.4
	CO concentration	Measured value	ppm	1100	1400
		O <sub>2</sub> - reduced value	ppm • 12%	3500	4200
gas analysis	NOx concentration	Measured value	ppm	10	14
		O <sub>2</sub> - reduced value	ppm • 12%	32	42
	SOx concentration	Measured value	ppm	21	15
		Amount emitted	m³ <sub>N</sub> /h	0.0000013	0.0000009
		Measured value	mg/m³ <sub>N</sub>	3300	260
	HUI CONCENTRATION	O <sub>2</sub> - reduced value	mg/m³ <sub>N</sub> •12%	11000	780

TABLE 10: Analysis results of emission gas properties.

Temperature of gas in the furnace; concentrations of dust, CO, NOx, SOx, and HCI measured 20 min after sample insertion.

Moisture content measured 5 min after sample insertion.

SOx emission calculated on the basis of 0.06 m3N/h of the amount of emitted gas.

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