Heavy Metals Recovery System for Dust from NKK Electric-resistance Furnace

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After vitrification of the residues from a municipal solid waste incinerator, the vitrified products can be used as a construction material, and the dust can be used as raw materials for the non-ferrous industry. The NKK electric-resistance furnace is operated under a reducing atmosphere, and heavy metals such as lead and zinc in the ash vaporize to form dust. The concentration of lead and zinc in the dust after washing is sufficiently high for the dust to be used as a raw material for the lead and zinc industry.

1. Introduction

Vitrification is considered a principal process for future waste disposal because the treatment can render harmless even wastes that include harmful substances, and it can recover resources in the waste. NKK has commercialized an electric-resistance ash melting furnace[1),2)] for processing residues from solid waste incineration plants, a high temperature gasifying and direct melting furnace[3)] for processing wastes, and a swirl flow melting furnace[4)] for treating sewage and sewage bottom ash that use vitrification technology developed by NKK over many years.

Residues from solid waste incineration plants are separated into slag, metal, and dust by vitrification treatment and then discharged. The metal is utilized for weights, while the dust is discarded in landfills after harmful metals in the dust are stabilized. The dust contains concentrated heavy metals such as Zn, Pb and Cd that were contained in the residues, so the present disposal method, in which the dust is stored and accumulated in a disposal area, is undesirable considering environmental preservation and resource recycling. Therefore, the dust should be reclaimed as a raw material for smelting.

The NKK electric-resistance ash melting furnace can volatilize the zinc and lead in residues at high rates using reduction melting and then concentrate these metals in the dust. NKK developed a heavy metal recovery process in which zinc and lead in the dust are reclaimed as a raw material for smelting by making the best use of the characteristics of the dust. Also, NKK proposed a system for reusing heavy metals as resources by incorporating a smelting process for zinc and lead in the heavy metal recovery process and demonstrated its practicality. Further, we assumed specific resource recovery system treatment routes and then estimated the energy consumption and CO_2 emissions for resource recovery treatment[5)].

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2. Metal content in residues from municipal solid waste incineration plants

About 50 million tons/year of waste is generated in Japan. 77% (in fiscal 1996) of this quantity is incinerated, generating about 6 million tons/year of bottom ash and fly ash residues. Bottom ash is an ember, and fly ash is the incinerator gas dust.

Table 1 gives an example[6] of chemical compositions of bottom ash and fly ash. Bottom ash contains a large quantity of metallic elements with a high boiling point, such as Fe, Cu and Mn. The fly ash is composed of a condensate of volatile components from the incineration process and ash in the incinerator gas. Fly ash contains a large amount of low boiling point metallic elements, such as zinc, lead and cadmium, and salt elements such as sodium, potassium and chlorine.

Table 1 Chemical composition of residues from MSW (Municipal Solid Waste) incineration plant

Element	Bottom ash mg/kg-ash	Fly ash mg/kg-ash
Si	165000	96600
Al	102000	64300
Ca	114000	120000
Fe	78500	21600
Mn	2400	1200
Mg	11400	14100
Na	19000	78700
K	7600	60000
Cl	15800	131000
Pb	2780	12600
Zn	5410	41800
Cd	23	466
Cu	3210	1490
Cr	350	230

The content of metallic elements in the bottom and fly ash generated from 1 ton of solid waste was estimated on the basis of chemical composition data[7] for bottom ash and fly ash obtained through questionnaires at 57 municipal solid waste incineration plants in Japan. The result is given in **Table 2**. It was estimated that the 160 kg of residue (134 kg of bottom ash + 26 kg of fly ash) from 1 ton of solid waste contain 480 g of zinc and 140 g of lead.

Table 3 gives the calculated amount of zinc and lead in the 6 million tons/year of residue generated in Japan, which was obtained from **Table 2**. The amount of zinc in the residue from municipal solid waste incineration plants in Japan is calculated to be 18000 tons/year. This corresponds to 2.9% of the 620000 tons/year[8] demand for zinc in fiscal 1997. Also, the amount of lead in the residue from municipal solid waste incineration plants in Japan is calculated to be 5300 tons/year, which corresponds to 2.0% of the 270000 tons/year[8] demand for lead.

Table 3 Chemical composition of residue from MSW incineration plant

	Demand in Japan (1997)	Amount of H.M. in MSW	
	t/year	t/year	Ratio
Zn	620000	18000	2.9 %
Pb	270000	5300	2.0 %

H.M. (Heavy Metal)

3. Vitrifying treatment and recovery of heavy metals

The elements of residues from municipal solid waste incineration plants are separated into slag, metal,

Table 2 Average content of metallic elements in residues from MSW incineration plants

Element	Concentration		Amount per t-MSW				
	Bottom ash	Fly ash	Bottom ash 134kg		Fly ash 26kg		Total 160kg
	g/t-ash	g/t-ash	kg/t-MSW	%	kg/t-MSW	%	kg/t-MSW
Si	174000	107000	23.3	89	2.78	11	26.1
Ca	140000	115000	18.8	86	2.99	14	21.8
Al	75000	74000	10.1	84	1.92	16	12.0
Fe	50700	13500	6.79	95	0.351	5	7.14
K	20400	68900	2.73	60	1.79	40	4.52
Na	20400	53500	2.73	66	1.39	34	4.12
Ti	8700	12100	1.17	79	0.315	21	1.48
Mg	-	23000	-	-	0.598	-	-
Zn	1130	12700	0.151	31	0.330	69	0.481
Pb	500	2790	0.067	48	0.073	52	0.140
Cu	909	477	0.122	91	0.012	9	0.134
Mn	597	610	0.080	83	0.016	17	0.096
Fe	140	870	0.019	45	0.023	55	0.042
Cr	128	190	0.017	77	0.0049	23	0.022
Cd	4	140	0.00051	12	0.0036	88	0.0041
Ni	27	-	0.0036	-	-	-	-
As	5	18	0.00062	57	0.00047	43	0.0011
Hg	0.03	3	0.000004	5	0.00008	95	0.00008

and dust by vitrifying treatment. Heavy metals, low boiling point elements, such as zinc, lead and cadmium, are concentrated in the dust.

At present, most of the dust is disposed of in landfills after toxic metals in the dust are stabilized. If the dust containing the heavy metals is used as a raw material[8], the "recycling of heavy metals in residues from municipal solid waste incineration plants" shown in **Fig. 1** becomes possible. Also, disposal of the dust in landfills is not needed, and the "zero emission treatment of residues from municipal solid waste incineration plants" shown in **Fig. 2** can be achieved.

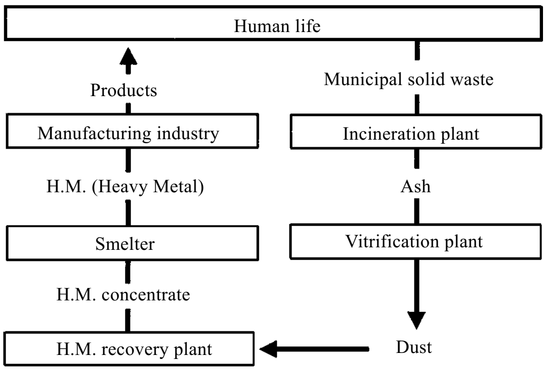


Fig. 1 Heavy metal recycling for MSW

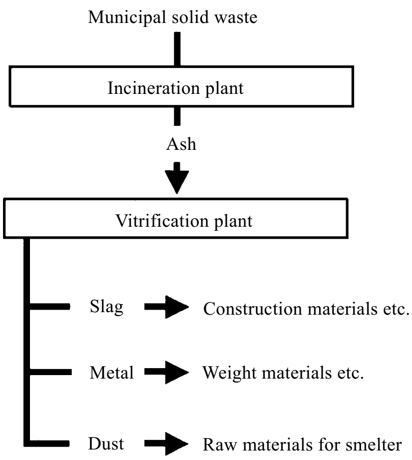


Fig. 2 Zero emission treatment for MSW

4. Generation of dust

Dust is composed of a concentrate of components that volatilize in the melting furnace, ash scattered from the residues supplied to the melting furnace, and ash entrained in the exhaust gas. Slaked lime is sometimes blown into oxidizing atmosphere furnace melting systems to remove the generated hydrogen chloride[9]. The reaction product and unreacted slaked lime currently migrate to dust. The composition, compound form, and amount of dust generated are affected by the melting process (e.g., melting system, furnace construction, atmosphere, temperature, and exhaust gas treatment method) and the composition of the supplied ash[9]. The components of the dust are as follows:

(1) Concentrates of volatile component: Na, K, Cl, Zn, Cd, Pb, etc.

(2) Ash scattered from the residues supplied to the melting furnace: Si, Ca, Al etc.

(3) Ash entrained in the exhaust gas: Si, Ca, Al etc.

(4) Product from slaked line injection: Ca, Cl, etc.

5. Dust of NKK electric-resistance ash melting furnace

5.1 Melting furnace construction and dust

The construction of the NKK electric-resistance ash melting furnace[1] is shown in **Fig. 3**. This electric-resistance ash melting furnace uses a system in which electric current flows through molten slag in the furnace, and incineration residues are melted by the generated resistance heat. The incineration residues supplied to the furnace float on the molten slag layer, and melt gently into the molten slag, so that the amount of ash scattered is small, compared to the other ash melting methods. Also, an electric furnace is used, so the amount of exhaust gas generated is small, and the amount of ash entrained in the exhaust gas is also small. The melting furnace has an enclosed construction, and the interior of the furnace is maintained in a reducing atmosphere. Therefore, zinc is reduced in the incineration residue and volatizes as metallic Zn. The volatizing metallic Zn is oxidized into ZnO in the discharge process. Also, very little hydrogen chloride is generated, so that slaked lime is not necessary. Since the molten slag layer is kept at a temperature of 1400–1500℃, components with a high boiling point such as Si do not volatize. Features of the NKK electric-resis-

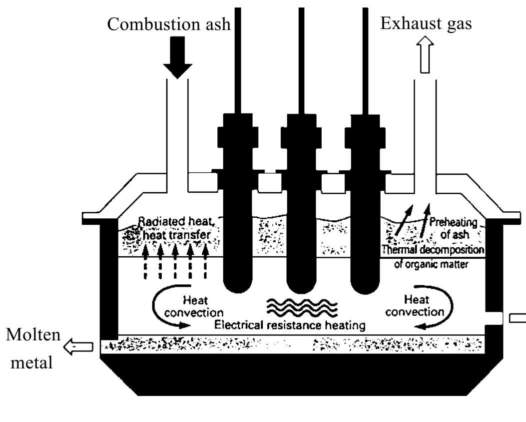


Fig. 3 NKK electric-resistance furnace

tance ash melting furnace concerning the generation of dust are as follows:

(1) Gentle melting: The amount of fly ash is small.

(2) Less exhaust gas: The amount of ash entrained in the exhaust gas is small.

(3) Reduction melting: Zn volatizes at high rates

(4) Less generated hydrogen chloride: Slaked lime need not to be blown in.

(5) Melting at 1500℃: Si does not volatize.

5.2 Migration behavior of metallic elements

An example[10) of material balance for the metallic elements in the NKK electric-resistance ash melting furnace is given in **Table 4**. 100% of the cadmium and 90% of the zinc and lead migrated to dust. Most of the iron and copper migrate to metal, and most of the silicon, aluminum, calcium, titanium, manganese and magnesium migrated to slag. In the NKK electric-resistance ash melting furnace, cadmium, zinc and lead migrate to dust at high rates.

Table 4 Material balance of metallic elements

unit : %

Element	Dust	Metal	Slag
Cd	100	0.0	0.0
Zn	90.4	5.6	4.0
Pb	90.3	9.6	0.1
Fe	0.13	93.2	6.7
Cu	0.19	98.8	1.0
Cr	0.07	60.6	39.3
Si	0.06	3.7	96.2
Al	0.04	0.02	100
Ca	0.04	0.04	100
Ti	0.12	3.1	96.8
Mn	0.17	9.9	89.9
Mg	0.04	0.03	100

5.3 Chemical composition of dust

An example[11) of the chemical composition of dust from the NKK electric-resistance ash melting furnace is given in **Table 5**. Although the component concentration of dust differed slightly between single melting of bottom ash and mixed melting of bottom ash and fly ash, the principal elements in the dust were zinc, lead, sodium, potassium and chlorine, and the total content of these elements was 85–95%. The zinc content was in the range of 20–30%, and the lead content was in the range of 6–10%. The total content of silicon, calcium and aluminum, which are ash elements, was less than 2%.

The dust generated from mixed melting of bottom ash and fly ash contains larger amounts of sodium, potassium and chlorine and smaller amounts of zinc and lead than the dust generated from single melting of the bottom ash. The reason for this is that the high concentrations of sodium, potassium and chlorine in the fly ash migrate to dust in large amounts.

Table 5 Chemical composition of dust from MF (Melting Furnace)

unit : %

Element	B.A.	B.A.+F.A.
Zn	27.0	21.4
Pb	10.0	6.4
Cd	0.10	0.31
Cu	0.11	0.14
Na	12.3	16.1
K	9.4	13.0
Cl	27.6	38.8
Si	0.23	0.57
Al	0.26	0.23
Ca	0.37	0.16

B.A.(Bottom Ash), F.A.(Fly Ash)

Fig. 4 shows an X-ray diffraction pattern[11) of the dust. NaCl, KCl and ZnO were confirmed as the principal compounds of the dust. The lead compounds were presumed to be PbO and $PbCl_2$[11) from the EDX (Energy Dispersive X-ray spectrometer) analysis. Also, electron microscopy suggested that the dust was an aggregate of fine particles with a particle size of about 1 μm[11).

Thus, dust from the NKK electric-resistance ash melting furnace is composed of NaCl, KCl, ZnO, PbO and $PbCl_2$, which are volatile components from the melting furnace, small amounts of ash elements such as Si, Ca and Al, and high concentrations of Zn and Pb.

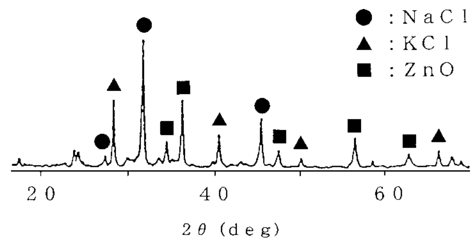


Fig. 4 X-ray diffraction pattern of dust from MF

6. Resource recovery system for dust

6.1 Separation of heavy metals in electric-resistance ash melting furnace

In the NKK electric-resistance ash melting furnace, zinc, cadmium and lead can be volatized at high rates of 90–100% based on the principle of reduction melting and can be concentrated in the dust. Also, little dust is generated because the incineration residue is floated on the molten slag layer and melted gently into the molten slag, and scattering of the incineration residue into the exhaust gas is restrained. Therefore, the dust scarcely contains silicon, calcium and aluminum, which are ash elements.

Thus, cadmium, zinc and lead in incineration residue can be separated from the dust at high rates by the unique melting principle and melting system of the NKK electric-resistance ash melting furnace.

6.2 Heavy metal recovery process

As shown in **Fig. 5**, dust from the electric-resistance ash melting furnace is composed of NaCl and KCl, which are water soluble components, and ZnO, PbO and $PbCl_2$, which are water insoluble components. Therefore, NaCl and KCl are extracted by washing treatment of the dust, and Zn and Pb concentrate can be obtained as residue[12].

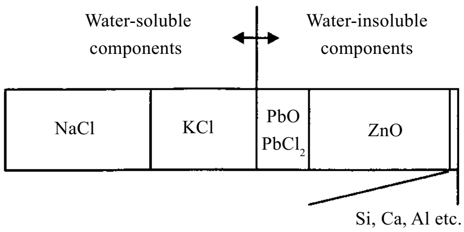


Fig. 5 Chemical components of dust from MF

Fig. 6 shows the heavy metal recovery process for dust from the melting furnace. The process includes washing treatment, solid-liquid separation treatment, and cleaning treatment by solvent. **Table 6** gives the chemical composition of heavy metal concentrate obtained by washing treatment and by heating after washing. Dust subjected to washing treatment is a zinc and lead concentrate that additionally contains small amounts of copper, tin and cadmium.

The zinc and lead concentrate can be used as a raw material for ISP (Imperial Smelting Process) smelting, which is a process for simultaneously smelting Zn and Pb. Although the allowable quality of raw material for ISP smelting is determined by the individual smelting plants, a minimum zinc content of 35% and a maximum chlorine content of 1% have been identified as an example quality[13].

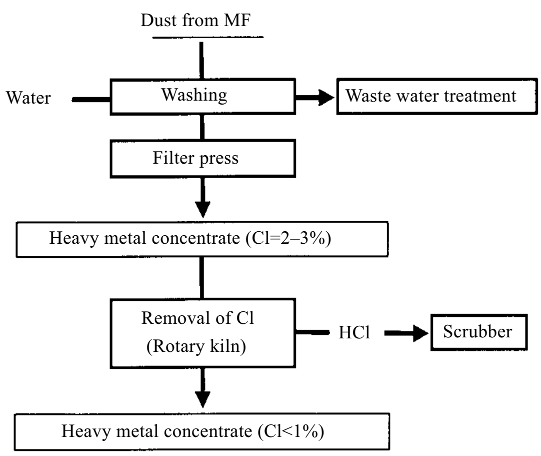


Fig. 6 Heavy metal recovery process for dust from MF

The dust subjected to washing treatment as specified in **Table 6** contains 48% zinc, which meets the allowable quality for a raw material for smelting. The chlorine content of 2–3% can be reduced to 0.5% or less by heating at a temperature of at least 800℃, although there are possible restrictions. The chlorine content was reduced to 0.4% by heating at 800℃ and to 0.01% and below at 900℃. Features of the heavy metal recovery process are as follows:

(1) Equipment configuration is simplified by the use of a washing treatment.

(2) Disposal of residues is unnecessary because washed residues are reused as resources.

Table 6 Chemical composition of heavy metal concentrate

unit : %

Element		Zn	Pb	Cl	Na	K	Cu	Sn
Dust from MF		21.4	5.9	29.8	13.2	12.9	0.20	0.32
Washing treatment		48.3	7.3	2.6	0.56	0.12	0.26	0.54
Heating treatment	800℃	54.8	10.9	0.44	0.65	0.12	0.41	0.82
	900℃	56.6	11.7	<0.01	0.67	0.13	0.51	0.94

6.3 Combination of heavy metal recovery process and ISP

In the ISP smelting process, oxides of Zn and Pb (sintered ore) are reduced with coke in a blast furnace to manufacture metallic Zn and metallic Pb[14]. The metallic zinc is recovered by cooling and condensing zinc vapor volatized in the blast furnace, while the metallic lead is recovered from the bottom of the blast furnace. At present, ISP plants are in operation at Hachinohe Refinery and Harima Refinery[13].

Zinc and lead concentrate obtained by the washing plus heating treatment of dust from the NKK electric-resistance ash melting furnace can be supplied to the ISP plant to reclaim metallic zinc and lead from the dust.

By combining the heavy metal recovery process with the ISP, the Zn and Pb metal separation step in the heavy metal recovery process can be omitted, simplifying the treatment. Also, dust from the NKK electric-resistance ash melting furnace contains very little scattered ash and ash entrained in the exhaust gas. Moreover, the dust is composed of water insoluble zinc and lead components and water soluble NaCl and KCl, so that a washing treatment can be used in the heavy metal recovery process.

7. Recycling of heavy metal resources

7.1 Material balance of Zn and Pb in incineration & vitrification plant

Heavy metals contained in solid waste are concentrated in incineration residues by the incineration treatment and are then concentrated further in the dust by vitrification treatment. A material balance for zinc and lead in an incineration and vitrification plant with a refuse throughput of 600 tons/day was calculated on the basis of **Tables 1** and **4**. The result is shown in **Fig. 7**. The amount of zinc reclaimed by supplying Zn and Pb concentrate recovered from dust to an ISP plant was calculated to be 95 tons/year, and the amount of lead reclaimed was calculated to be 28 tons/year.

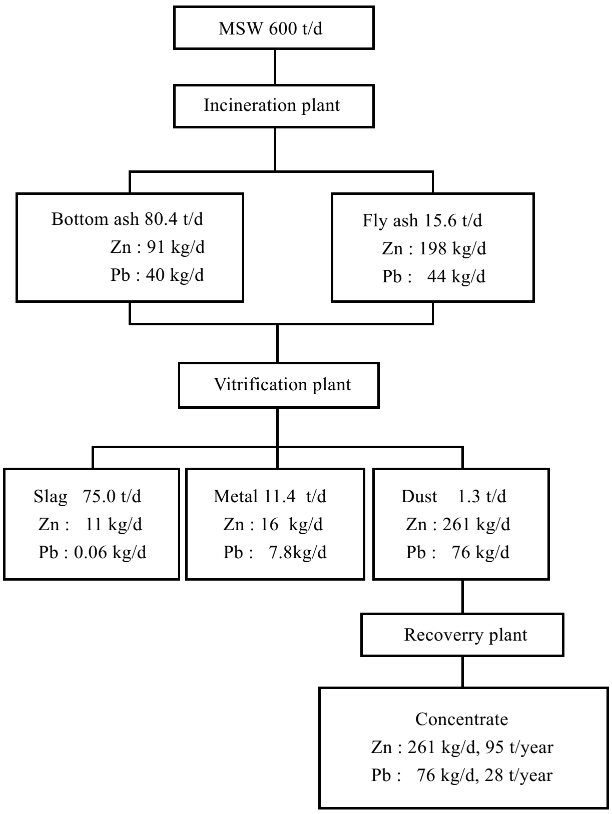


Fig. 7 Example of material balance of Zn and Pb for MSW treatment plant

7.2 Treatment route for resource recovery of dust

The resource recovery treatment of dust (washing treatment plus heating treatment) could be accomplished in a vitrification plant, centralized treatment plant, smelting plant, or similar facility.

As an example, we assumed three routes (R1, R2 and R3) for supplying dust generated in a vitrification plant in city A in the Kanagawa Prefecture to a smelting plant in city B in the Aomori Prefecture (travel distance: 730 km) after the dust was treated for resource recovery, as shown in **Fig. 8**. The energy consumption and CO_2 emission for these three routes were evaluated[5).

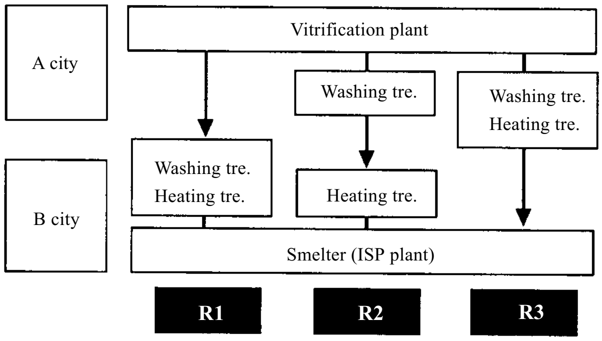


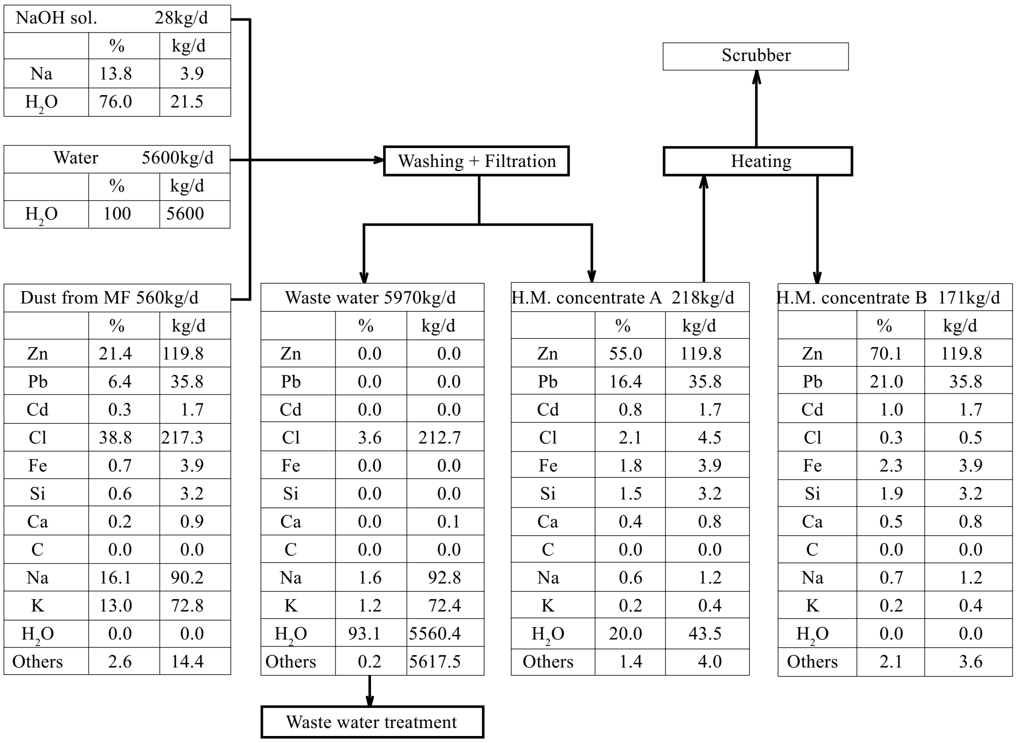
Fig. 8 Routes for heavy metal recycling system

The condition was set based on prototype test data[1) for the NKK electric-resistance ash melting furnace: 28 tons/day of incineration residues generated from the incineration of 200 tons/day of refuse are vitrified, and the 560 kg/day (204 tons/year) of generated dust is treated for resource recovery. It was assumed that the chemical composition of the dust was the same as for mixed melting of bottom ash and fly ash shown in **Table 6**. **Fig. 9** shows an example material balance for resource recovery treatment of 560 kg of dust. It was also assumed that all of the washing treatment equipment and heating treatment equipment required for resource recovery were newly built, and the calculations included the energy consumption and CO_2 emission required for building the equipment. In the smelting plant, the amount of decrease of the energy consumption and CO_2 emission required for ore transportation and the like that were saved by the supply of Zn and Pb concentrate recovered from dust were considered as saved amounts.

Table 7 and **Fig. 10** show the energy consumption and CO_2 emission required for transportation (on land), washing treatment, heating treatment, and smelting in the resource recovery treatment of dust. The energy consumption and CO_2 emission increase in the order of R1, R2 and R3, with route R1 showing the best result. Each route is described below:

(1) Washing and heating treatment in smelting plant (R1)

Since the volume of transportation is higher than that of R2 and R3, the energy consumption and CO_2 emission for transportation are high. However, since washing treatment and (rotary kiln) heating treatment are conducted using large equipment in the smelting plant, advantages of scale are important, so that the



NaOH sol. 28kg/d	%	kg/d
Na	13.8	3.9
H_2O	76.0	21.5

Water 5600kg/d	%	kg/d
H_2O	100	5600

	Dust from MF 560kg/d		Waste water 5970kg/d		H.M. concentrate A 218kg/d		H.M. concentrate B 171kg/d	
	%	kg/d	%	kg/d	%	kg/d	%	kg/d
Zn	21.4	119.8	0.0	0.0	55.0	119.8	70.1	119.8
Pb	6.4	35.8	0.0	0.0	16.4	35.8	21.0	35.8
Cd	0.3	1.7	0.0	0.0	0.8	1.7	1.0	1.7
Cl	38.8	217.3	3.6	212.7	2.1	4.5	0.3	0.5
Fe	0.7	3.9	0.0	0.0	1.8	3.9	2.3	3.9
Si	0.6	3.2	0.0	0.0	1.5	3.2	1.9	3.2
Ca	0.2	0.9	0.0	0.1	0.4	0.8	0.5	0.8
C	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Na	16.1	90.2	1.6	92.8	0.6	1.2	0.7	1.2
K	13.0	72.8	1.2	72.4	0.2	0.4	0.2	0.4
H_2O	0.0	0.0	93.1	5560.4	20.0	43.5	0.0	0.0
Others	2.6	14.4	0.2	5617.5	1.4	4.0	2.1	3.6

Fig. 9 Example of material balance for heavy metal recovery plant

Table 7 Calculated energy consumption and CO_2 emission for recycling routes

Route	Location	Transport Treatment Smelting	Amount of treatment t/year	Energy consumption GJ/year	CO_2 emission kg-C/year
R1	A→B	Transport	204.4	164	3074
	B city	Washing tre.	204.4	209	2889
	B city	Heating tre.	79.5	457	7262
	B city	Smelting	62.1	–223	–6
			Total	607	13219
R2	A city	Washing tre.	204.4	1003	15005
	A→B	Transport	79.5	64	1196
	B city	Heating tre.	79.5	457	7262
	B city	Smelting	62.1	–223	–6
			Total	1301	23457
R3	A city	Washing tre.	204.4	2079	29986
	A city	Heating tre.	79.5		
	A→B	Transport	62.1	50	934
	B city	Smelting	62.1	–223	–6
			Total	1907	30914

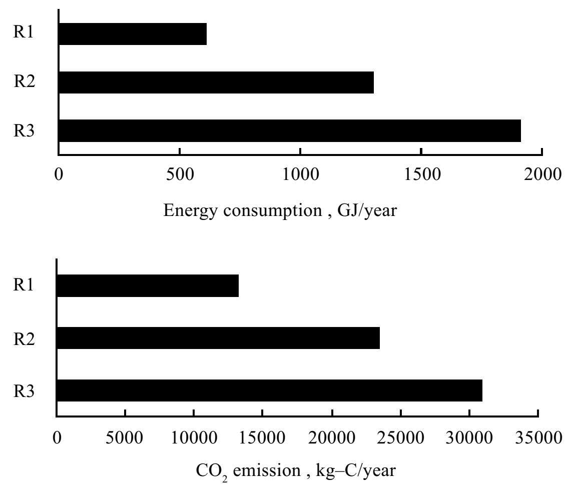


Fig. 10 Calculated energy consumption and CO_2 emission for recycling routes

energy consumption and CO_2 emission are the lowest.

(2) Washing treatment done in vitrification plant and heating treatment in smelting plant (R2)

The energy consumption and CO_2 emission take intermediate values between R1 and R3, and are about two times those of R1.

(3) Washing and heating treatment in vitrification plant (R3)

The volume of transportation is low, and therefore the energy consumption and CO_2 emission in transportation are low. However, since washing treatment and (electrical) heating treatment are conducted using small equipment at the vitrification plant, the energy consumption and CO_2 emission are the highest, being about three times those of R1.

Thus, significant saving of the energy consumption and CO_2 emission required for resource recovery treatment of dust should be possible by utilizing large equipment in the smelting plant[5]. In determining the route for resource recovery treatment of dust, the selection of a rational and economical route is desired, although many restrictions are expected.

8. Conclusion

Heavy metals in solid waste are concentrated in dust during vitrification. The dust is a harmful waste if it is discarded, but a heavy metal resource if recovered. Although the amount of dust generated is small, the reuse of dust as a raw material for smelting is of significance. The most valuable function of the vitrification of incineration residue is to render it harmless. The separation of harmful metals for recovery is superior to other stabilizing treatments.

By reusing dust from vitrification as a raw material for smelting, the incineration plus vitrification treatment is made a "refuse disposal system suitable for environment preservation" that achieves both harmlessness and resource recovery. Also, the zero emission treatment of residues from municipal solid waste incineration plants is attained, and the recycling of heavy metals discharged into solid waste is completed.

The NKK electric-resistance ash melting furnace provides high performance in separating heavy metals and can produce dust advantageous for reuse as a raw material for smelting and safe slag containing less heavy metals. This system can ideally fulfil the function of vitrification.

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