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# Treatment of hazardous sorbents generated from the adsorption of heavy metals during incineration

Ming-Yen Wey\*, Ming-Horng Yan, Jyh-Cherng Chen

Department of Environmental Engineering, Chung-Hsing University, Taichung 402, Taiwan, ROC

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#### Abstract

The emission of heavy metals during waste incineration can be effectively reduced through the practice of employing non-toxic sorbents. These sorbents can react with toxic metals at high temperatures and create metal binding between them by various physical and chemical mechanisms. After the adsorption process, the used sorbents, which contain heavy metals, need to be desorbed to reduce their potential environmental hazards or provide reusable sorbents for economical aspect. The sorbent's adsorption efficiency is affected by different operating conditions and waste elemental compositions during incineration, which, in turn, affect their desorption characteristics. However, the effects of operating condition and waste elemental composition on the stability of heavy metals in the sorbents and the desorption efficiencies have been little studied. This study investigates the desorption characteristics of heavy metals (Cr, Pb, Cu, and Cd) from the hazardous sorbents with different extracting reagents (H<sub>2</sub>O, HCl, EDTA, and Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub>). The hazardous sorbents were generated under different adsorption time and various input waste elemental compositions during incineration process. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: Adsorption; Sorbent; Extracting agents; HCl; EDTA; Na2S2O5

# 1. Introduction

With the advantages of toxicity destruction, waste volume reduction, and potential energy recovery, incineration has become the prevalent treatment of solid waste in Taiwan. However, the flue gas, waste water and residues from incineration always

<sup>\*</sup> Corresponding author.

pollute the environment due to improper operation, complex waste and unclassified waste. The emission of heavy metals during incineration and the leaching of heavy metals from ash have received considerable attention recently.

Toxic metal compounds in waste cannot be destroyed and will volatilize during incineration and subsequently condense to form metallic particles during the cooling of flue gas. These metallic particles are submicron in size, which may not always be effectively collected by conventional air pollution control system. Those particles are hazardous to both human health and the surrounding environment.

Previous studies of heavy metal control [1-3] indicate that it is difficult and uneconomic to control the volatilization of metals during high-temperature processes. An alternative control technology for heavy-metal emission is using solid sorbents to capture metals by physical deposition and chemical adsorption. Among available incineration systems, the fluidized bed incinerator is best suited for this purpose, because it provides intensive mixing of sorbents and metallic compounds.

The adsorption mechanisms of sorbents to capture metals during incineration include: (1) metallic ash capture: some unvolatilized metals would either remain in metallic ash and be captured by sorbents or else be deposited; this mechanism is sensitive to operating temperature; (2) vapor capture: some volatilized metals exist in gaseous phase, and then captured by sorbents through homogeneous nucleation, heterogeneous deposition, and chemical adsorption. This mechanism is particularly effective as smaller sorbent is used; (c) particulate capture: most metallic particulates can be captured by sorbents through processes of coagulation, Brownian motion, interception, and impaction. This is particularly evident for the sorbent with sticky surface, e.g. glassy surface or sorbents coated with a layer of stick material [2,3].

After incineration, the metallic compounds which did not volatilize during incineration will exist in bottom ash. However, the concentration of heavy metals in bottom ash is greater than that in waste due to volume reduction. Thus, the concentration of heavy metals in sorbents must be higher when they are used to control the emission of heavy metals during incineration. These sorbents need to be treated carefully to prevent secondary pollution. These hazardous materials are usually treated by solidification or stabilization then disposed to a landfill, or even directly disposed to a landfill. The leaching water from the landfills will slowly pollute the water or underground water in the environment [4]. Although the solidification can inhibit the leaching tendency of pollutants, it still has the disadvantages of increasing waste volume and doubtful long-term stability. Thus, many experts devoted to the study of reusing these materials have found that the leaching of heavy metals is a major obstacle. In order to reduce the environmental risk, the sorbents can be treated by extracting methods which can remove the heavy metals from the sorbents and reduce its hazardous nature. Acidic aqueous solutions, basic solutions, and surfactant are conventionally employed to extract heavy metals from the sorbents. The desorption mechanisms of extracting agents during extracting processes include: (1) for HCl, heavy metals are removed by ion exchange at low pH; (2) for EDTA, heavy metals are removed by chelation; (3) for  $Na_2S_2O_5$ , heavy metals are removed by reduction reaction [5-7].

In order to reduce the potential environmental hazards of these sorbents or provide reusable materials by lowering the concentration of heavy metals, the extracting methods which are usually used in soil remediation processes [5-11] are carried out to dispose of the sorbents (silica sand mixed with limestone) generated from different incineration conditions. The evaluated parameters in this study include: (1) determining the optimal operating concentration of different extracting agents; (2) comparing the extracting efficiencies of heavy metals with different extracting agents (H<sub>2</sub>O, HCl, EDTA, and Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub>); and (3) the influence of different incineration conditions on the extracting efficiencies of heavy metals for each extracting agent.

# 2. Experimental

#### 2.1. Properties of the sorbents

The sorbents used in this study were generated from various incineration conditions from different adsorption times and different input waste elemental compositions. Tables 1 and 2 list the corresponding incineration conditions and heavy metal concentrations.

# 2.2. Experimental procedure

The experiment was carried out in two stages. The optimal operating concentrations of different extracting agents were determined in the first stage, and the effects of different incineration conditions on the extracting efficiencies of four heavy metals by different extracting agents were investigated in the second stage. The standard derivation of the extracting efficiency is about 0.5%-4%. The detailed procedure of each stage is described below.

# 2.3. The optimal operating concentration of different extracting agents

The sorbents that have adsorbed heavy metals under various adsorption time and input waste elemental compositions were the major materials to be desorbed in this study. When the input waste did not contain chloride, Samples 3, 6, 9, and 12 were chosen. In the same way, Samples 14, 18, 20, and 24 were chosen for polyvinyl chloride (PVC) additives, and Samples 27, 30, 33, and 36 were chosen for NaCl additives. The extracting agents and their concentrations were HCl (0, 0.1, 0.4, 0.7, and 1.0 M), EDTA (0, 0.05, 0.075, 0.10, and 0.125 M), and Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub> (0, 0.1, 0.4, 0.7, and 1.0 M). In the extracting agent was added. The beaker revolved for 24 h at a speed of 180 rpm using a mechanical shaker. The pH value of each sample was measured and recorded. Each sample was then filtered by a filter with 0.45  $\mu$ m pore size. Finally, the concentrations of heavy metals in the filtrates were analyzed using an atomic absorption spectrophotometer. These analyzed values represented the total amount of heavy metals

Number	Chloride	Incineration	Adsorption	Heavy metal	pH <sup>a</sup>
	species temperature (°C) time (min		time (min)	(mg/g)	1
1	_	700	30	Cr: 1.42	12.25
2	-	700	60	Cr: 2.26	12.24
3	-	700	90	Cr: 4.86	12.27
4	-	700	30	Pb: 5.70	12.25
5	-	700	60	Pb: 8.38	12.24
6	-	700	90	Pb: 13.29	12.24
7	-	700	30	Cu: 3.16	12.25
8	-	700	60	Cu: 6.92	12.23
9	_	700	90	Cu: 10.70	12.21
10	-	700	30	Cd: 0.92	12.21
11	_	700	60	Cd: 1.28	12.23
12	-	700	90	Cd: 1.48	12.25
13	PVC	700	30	Cr: 6.39	12.06
14	PVC	700	60	Cr: 11.06	12.10
15	PVC	700	90	Cr: 6.65	12.09
16	PVC	700	30	Pb: 1.47	12.19
17	PVC	700	60	Pb: 3.43	12.10
18	PVC	700	90	Pb: 5.16	12.16
19	PVC	700	30	Cu: 4.00	12.12
20	PVC	700	60	Cu: 6.50	12.08
21	PVC	700	90	Cu: 4.00	12.14
22	PVC	700	30	Cd: 0.30	12.14
23	PVC	700	60	Cd: 0.30	12.09
24	PVC	700	90	Cd: 0.85	12.13
25	NaCl	700	30	Cr: 5.70	12.22
26	NaCl	700	60	Cr: 11.20	12.22
27	NaCl	700	90	Cr: 13.10	12.23
28	NaCl	700	30	Pb: 2.62	12.26
29	NaCl	700	60	Pb: 5.04	12.27
30	NaCl	700	90	Pb: 10.65	12.26
31	NaCl	700	30	Cu: 6.02	12.25
32	NaCl	700	60	Cu: 16.50	12.25
33	NaCl	700	90	Cu: 17.60	12.21
34	NaCl	700	30	Cd: 1.65	12.25
35	NaCl	700	60	Cd: 2.05	12.22
36	NaCl	700	90	Cd: 3.15	12.26

Sorbents generated from different conditions and their pH and heavy metal concentration (1)

<sup>a</sup>The pH in 0.01 M CaCl<sub>2</sub> solution.

released/leached from the sorbents. The extracting efficiency of each test was calculated from the formula:

Extracting Efficiency 
$$R(\%) = \left\{ \frac{W_{\rm f}}{W_{\rm s}} \right\} \times 100\%$$
,

where  $W_{\rm f}$  = the amount of heavy metal in the filtrate and  $W_{\rm s}$  = the amount of heavy metal in the sorbent.

Table 1

Number	Chloride	Incineration	Adsorption	Heavy metal	pH <sup>a</sup>
	species	temperature (°C)	time (min)	(mg/g)	r
1	_	700	30	Cr: 1.42	12.25
2	_	700	60	Cr: 2.26	12.24
3	-	700	90	Cr: 4.86	12.27
4	-	700	30	Pb: 5.70	12.25
5	-	700	60	Pb: 8.38	12.24
6	-	700	90	Pb: 13.29	12.24
7	-	700	30	Cu: 3.16	12.25
8	-	700	60	Cu: 6.92	12.23
9	_	700	90	Cu: 10.70	12.21
10	-	700	30	Cd: 0.92	12.21
11	-	700	60	Cd: 1.28	12.23
12	-	700	90	Cd: 1.48	12.25
37	_	600	30	Cr: 0.61	12.26
38	_	600	60	Cr: 1.28	12.27
39	_	600	90	Cr: 2.16	12.25
40	_	600	30	Pb: 6.55	12.27
41	_	600	60	Pb: 15.02	12.26
42	_	600	90	Pb: 16.08	12.25
43	_	600	30	Cu: 4.34	12.25
44	_	600	60	Cu: 10.40	12.25
45	_	600	90	Cu: 10.86	12.25
46	_	600	30	Cd: 0.84	12.24
47	_	600	60	Cd: 1.21	12.23
48	_	600	90	Cd: 2.32	12.23
49	_	800	30	Cr: 2.66	12.26
50	_	800	60	Cr: 4.56	12.26
51	_	800	90	Cr: 5.55	12.25
52	_	800	30	Pb: 5.59	12.25
53	_	800	60	Pb: 10.23	12.24
54	_	800	90	Pb: 11.85	12.27
55	_	800	30	Cu: 4.74	12.26
56	_	800	60	Cu: 10.71	12.25
57	_	800	90	Cu: 18.72	12.25
58	_	800	30	Cd: 0.50	12.25
59	_	800	60	Cd: 0.88	12.24
60	-	800	90	Cd: 1.95	12.25

Table 2												
Sorbents	generated	from	different	conditions	and	their	pН	and	heavy	metal	concentratio	on (2)

<sup>a</sup>The pH in 0.01 M  $CaCl_2$  solution.

# 2.4. The effect of different incineration conditions on the extracting efficiencies of heavy metals

The sorbents used in this stage are all listed in Table 1, and the concentration of each extracting agent herein was the optimal operating concentration determined from the first stage experiment. The extracting procedure was the same as that described above.

#### 3. Results and discussion

### 3.1. The optimal operating concentration of different extracting agents

Figs. 1–3 show the effects of extracting agents concentrations on the extracting efficiencies of heavy metals under different input waste compositions. The results indicate that the optimal operating concentrations of HCl, EDTA, and  $Na_2S_2O_5$  are 0.4, 0.075, and 0.4 M, respectively. When the concentrations are higher than these values, they do not have significant effects on the extracting efficiencies of heavy metals. Figs. 1 and 3 show that HCl and  $Na_2S_2O_5$  have higher efficiencies in extracting Cu than other metals, while EDTA is better at extracting Cr as shown in Fig. 2. Among these extracting efficiencies of Cr, Cu, and Cd than that of EDTA in most cases, but the extracting efficiencies of  $Na_2S_2O_5$  on Pb are very low. It may be because  $Na_2S_2O_5$  is ineffective at extracting Pb or that  $Na_2S_2O_5$  reacts with Pb to form suspended chemical compounds which are difficult to remove from the filtrate.

3.2. Effects of different input waste compositions and adsorption time on the extracting efficiencies of heavy metals

#### 3.2.1. Extracting by deionized water

Fig. 4 reveals the effect of different input waste compositions and adsorption time on the extracting efficiencies of heavy metals by deionized water. In Fig. 4a, when the input waste does not contain chloride or NaCl, more than 50% of Cr in the sorbents can be removed by deionized water. Their extracting efficiencies are higher than that containing PVC at the three studied adsorption times. Perhaps soluble compounds of Cr are the major species during the incineration process. In Fig. 4b, when input waste contains NaCl, the extracting efficiency of Pb by deionized water is higher than that of feedstock containing PVC and no chloride, and increases as the adsorption time increases. As for Cu and Cd, no matter how long the adsorption time and how much the chloride is contained, the extracting efficiencies of Cu and Cd are zero. This reveals that the bindings between sorbents and the species of Cu and Cd are stronger than that of Cr and Pb.

#### 3.2.2. Extracting by 0.4 M HCl

Fig. 5 illustrates the effect of different input waste compositions and adsorption time on the extracting efficiencies of heavy metals by HCl. In Fig. 5a, when the adsorption time is shorter than 90 min, the presence of chloride decreases the extracting efficiency of Cr. When the adsorption time is 90 min, the effect of chloride on extracting Cr is not obvious. In Fig. 5b, the highest extracting efficiency of Pb occurs when the feed waste contains PVC and the adsorption time is 30 min. When the adsorption time is increased to 60 min and 90 min, the extracting efficiencies are the lowest as containing PVC. In Fig. 5c, when the adsorption time is longer than 30 min, the extracting efficiencies of Cu under different input waste compositions are in the order of containing no chloride > containing NaCl > containing PVC. When the input waste contains no chloride, the



Fig. 1. The effect of HCl concentration on extracting efficiencies under different input waste species: (a) without chloride; (b) containing PVC; (c) containing NaCl.

![](_page_7_Figure_1.jpeg)

Fig. 2. The effect of EDTA concentration on extracting efficiencies under different input waste species: (a) without chloride; (b) containing PVC; (c) containing NaCl.

![](_page_8_Figure_1.jpeg)

Fig. 3. The effect of  $Na_2S_2O_5$  concentration on extracting efficiencies under different input waste species: (a) without chloride; (b) containing PVC; (c) containing NaCl.

![](_page_9_Figure_1.jpeg)

Fig. 4. The effect of different input waste compositions and adsorption times on the extracting efficiencies of heavy metals by deionized water.

efficiency could reach as high as 94%. In Fig. 5d, the extracting efficiencies of Cd are independent of the adsorption time and in the order of containing no chloride > containing PVC > containing NaCl. Perhaps major metal compounds which form during incineration are metallic chloride in most case when the input contains chloride, so that HCl inhibits the heavy metal species from dissolving, as shown in Fig. 5.

#### 3.2.3. Extracting by 0.075 M EDTA

Fig. 6 shows the effect of different input waste compositions and adsorption times on the extracting efficiencies of heavy metals by EDTA. In Fig. 6a, when the adsorption time is shorter than 90 min, the extracting efficiencies of Cr follow the sequence of containing no chloride > containing NaCl > containing PVC. When the adsorption time reaches 90 min, the sequence becomes containing NaCl > containing PVC > containing no chloride. In Fig. 6b, when the input waste contains PVC, the highest extracting efficiency of Pb by EDTA occurs at the adsorption time of 30 min. When the adsorption time is longer than 30 min, the extracting efficiencies of Pb follow the sequence of

![](_page_10_Figure_1.jpeg)

Fig. 5. The effect of different input waste compositions and adsorption times on the extracting efficiencies of heavy metals by 0.4 M HCl.

![](_page_11_Figure_1.jpeg)

Fig. 6. The effect of different input waste compositions and adsorption times on the extracting efficiencies of heavy metals by 0.075 M EDTA.

containing NaCl > containing no chloride > containing PVC. In Fig. 6c, when the adsorption time is shorter than 90 min, the extracting efficiencies of Cu follow the sequence of containing no chloride > containing PVC > containing NaCl. Without chloride, the extracting efficiencies of Cu are the highest, no matter how long the adsorption time is. In Fig. 6d, when the adsorption time is longer than 30 min, the extracting efficiencies of Cd follow the sequence of containing no chloride > containing PVC > containing PVC > containing PVC > containing PVC > containing NaCl. The lowest extracting efficiency of Cd occurs when the adsorption time is 30 min and the input waste contains NaCl. When input waste contains NaCl, the exchangeable site on sorbent may already be held by Na<sup>+</sup> instead of by compounds of Cu and Cd during incineration, so that the ion exchange ability of Na<sup>+</sup> solution is reduced as shown in Fig. 6c and d.

# 3.2.4. Extracting by 0.4 M Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub>

Fig. 7 shows the effect of different input waste compositions and adsorption time on the extracting efficiencies of heavy metals by  $Na_2S_2O_5$ . In Fig. 7a, when the adsorption time is shorter than 90 min, the extracting efficiencies of Cr follow the sequence of containing no chloride > containing NaCl > containing PVC. The trend of extracting efficiencies of Cr by  $Na_2S_2O_5$  is similar to that of HCl. The extracting efficiencies of Pb by  $Na_2S_2O_5$  under different incinerating conditions are all zero. This result coincided with that in the test of optimal operating concentration of different extracting agents. That may be due to the fact that  $Na_2S_2O_5$  is ineffective in extracting Pb or because  $Na_2S_2O_5$  reacts with Pb on the sorbents to form suspended chemical compounds so that the concentration of Pb in the filtrate is quite low. In Fig. 7b and c, it is obvious that no matter how long the adsorption time is, the extracting efficiencies of Cu and Cd by  $Na_2S_2O_5$  follow the sequence of no chloride > containing PVC > containing NaCl. The trend in Fig. 7b and c is similar to Fig. 6c and d, for the reason described before.

# 3.3. Effects of different incineration temperatures and adsorption time on the extracting efficiencies of heavy metals

### 3.3.1. Extracting by deionized water

Fig. 8 reveals the effect of different incineration temperatures and adsorption time on the extracting efficiencies of heavy metals by deionized water. In Fig. 8a, when the incineration temperatures are 600°C and 700°C, the extracting efficiencies of Cr by deionized water are the best when the adsorption time is 60 min. The best extracting efficiencies of Cr by deionized water occur at 700°C under the three adsorption times. In Fig. 8b, it is obvious that the extracting efficiencies of Pb are lower than those of Cr, and the lowest efficiencies occur when the adsorption time is 60 min. When extracting Cu and Cd, no matter how long the adsorption time is and how much the chloride contained is, the extracting efficiencies of Cu and Cd are zero. Therefore, it is useless to decrease the amount of Cr and Cd in sorbents by extracting treatment with deionized water.

#### 3.3.2. Extracting by 0.4 M HCl

Fig. 9 illustrates the effect of different incineration temperatures and adsorption times on the extracting efficiencies of heavy metals by HCl. In Fig. 9a, when the incineration

![](_page_13_Figure_1.jpeg)

Fig. 7. The effect of different input waste compositions and adsorption times on the extracting efficiencies of heavy metals by 0.4 M  $Na_2S_2O_5$ .

temperature is 700°C, the extracting efficiencies of Cr by 0.4 M HCl are the best, and the extracting efficiency decreases as the adsorption time increases. In Fig. 9b, the extracting efficiency of Pb by 0.4 M HCl decreases as the incineration temperature or adsorption time increases. In Fig. 9c, the extracting efficiencies of Cu by 0.4 M HCl are high and the influence of incineration temperature on extracting efficiencies of Cu is not obvious when the adsorption time is 30 min. But when the incineration temperature is 800°C, the extracting efficiencies of Cu are lower than those at 600°C or 700°C when

![](_page_14_Figure_1.jpeg)

Fig. 8. The effect of different incineration temperatures and adsorption times on the extracting efficiencies of heavy metals by deionized water.

the adsorption time is longer than 30 min. In Fig. 9d, the extracting efficiencies of Cd are in the order of  $700^{\circ}$ C >  $600^{\circ}$ C >  $800^{\circ}$ C when the adsorption time is longer than 30 min.

#### 3.3.3. Extracting by 0.075 M EDTA

Fig. 10 shows the effect of different incineration temperatures and adsorption times on the extracting efficiencies of heavy metals by EDTA. In Fig. 10a, the highest extracting efficiency of Cr by 0.075 M EDTA occurs at 700°C and 30 min adsorption time. In Fig. 10b, the extracting efficiencies of Pb follow the sequence of  $600^{\circ}$ C >  $700^{\circ}$ C >  $800^{\circ}$ C, and the efficiency increases as the adsorption time decreases at  $600^{\circ}$ C or  $700^{\circ}$ C. In Fig. 10c, when the adsorption time is 30 or 90 min, the extracting efficiencies of Cu follow the sequence of  $600^{\circ}$ C >  $700^{\circ}$ C >  $800^{\circ}$ C, but the sequence becomes  $700^{\circ}$ C >  $600^{\circ}$ C >  $800^{\circ}$ C when the adsorption time is 60 min. In Fig. 10d, the influence of the adsorption time and incineration temperature upon the extracting efficiencies of Cd is not significant.

#### 3.3.4. Extracting by 0.4 $M Na_2S_2O_5$

Fig. 11 shows the effect of different incineration temperatures and adsorption times on the extracting efficiencies of heavy metals by  $Na_2S_2O_5$ . In Fig. 11a, the highest

![](_page_15_Figure_1.jpeg)

Fig. 9. The effect of different incineration temperatures and adsorption times on the extracting efficiencies of heavy metals by 0.4 M HCl.

![](_page_16_Figure_1.jpeg)

Fig. 10. The effect of different incineration temperatures and adsorption times on the extracting efficiencies of heavy metals by 0.075 M EDTA.

![](_page_17_Figure_1.jpeg)

Fig. 11. The effect of different incineration temperatures and adsorption times on the extracting efficiencies of heavy metals by 0.4 M  $Na_2S_2O_5$ .

extracting efficiency of Cr by 0.4 M Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub> occurs at 700°C and 30 min adsorption time. The trend of extracting efficiencies of Cr by Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub> is similar to those by HCl or EDTA, and this shows that the remaining Cr is difficult to remove by the extracting agents we used here. The extracting efficiencies of Pb by Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub> under different incinerating conditions are very low. This result coincides with that of the first stage test and the reason is the same. In Fig. 11b, the highest extracting efficiencies of Cd by Na<sub>2</sub>S<sub>2</sub>O<sub>5</sub> follow the sequence of 700°C > 600°C > 800°C when the adsorption time is 30 or 90 min.

## 4. Conclusions

From the above experimental results and discussion, the following conclusions can be drawn.

(1) The optimal operating concentrations of HCl, EDTA, and  $Na_2S_2O_5$  are 0.4, 0.075, and 0.4 M, respectively.

(2) HCl and  $Na_2S_2O_5$  have higher extracting efficiencies of Cu than the other metals, and EDTA has the highest extracting efficiencies of Cr.

(3) During the extraction process by deionized water, the extracting efficiencies of Cr were high. This means that most species of Cr formed during incineration are watersoluble, and may easily leach into the environment. These sorbents need to be treated with care.

(4) When the input waste contains chloride, the extracting efficiencies of Cr, Cu, and Cd by HCl, EDTA, and  $Na_2S_2O_5$  decrease in most cases.

(5) When the incineration temperature is 700°C, the extracting efficiencies of Cr by deionized water, HCl, EDTA, and  $Na_2S_2O_5$  are the highest. The highest extracting efficiencies of Pb by HCl and EDTA, and that of Cu by  $Na_2S_2O_5$  occurred at 600°C and 700°C, respectively.

(6) The influence of the adsorption time on extracting efficiencies of heavy metals by different extracting agents is complicated and relates to the incineration temperature, input waste composition and extracting agents.

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