

Release characteristics and water washing of heavy metals from electroplating sludge during low-temperature drying

Huaiyu Liu^{a,b}, Xiaoting Hong^{a,*}, Keming Wu^b, Kwan San Hui^c

^aDepartment of Chemistry, Zhejiang Sci-Tech University, Hangzhou 310018, China, Tel. +86 (0571) 86843228; Fax: +86 (0571) 86843228; email: hanren.xiaoting@gmail.com

^bHubei Key Laboratory for Efficient Utilization and Agglomeration of Metallurgic Mineral Resources, Wuhan University of Science and Technology, Wuhan 430081, China, Tel. +8615671559395; email: 601053136@qq.com (H.Y. Liu), Tel. +8613212730361; email: 82681936@qq.com (K.M. Wu)

^eEngineering, Faculty of Science, University of East Anglia, Norwich, NR4 7TJ, United Kingdom, email: k.hui@uea.ac.uk

Received 25 March 2021; Accepted 31 July 2021

ABSTRACT

This work demonstrates the electroplating sludge (ES) drying and the control method of heavy metal exhaust during the low-temperature drying process. The drying efficiency of ES has a maximum value at an air volume of 1.1 m³ h⁻¹ and a drying temperature of 70°C. The release amounts of heavy metals of the process were measured with inductively coupled plasma. Specifically, the release amounts were 0.009, 0.124, 0.170 and 2.097 μ g g⁻¹ for Cu, Ni, Mn and Zn without the employment of control methods, respectively, which exceed China's National Standard. Nevertheless, the release amount of heavy metals can meet the National Standard after the exhaust gas was washed by absorption solution with pH above 5. Further, the maximum absorption rates of Cu, Ni, Mn and Zn were 63.97%, 98.79%, 98.16% and 99.24%, respectively at a pH of 13. The absorption rates of Cu, Ni, Mi and Zn were 63.67%, 90.68%, 97.87% and 91.37%, respectively when water was used as the absorption solution with a pH of 7, which was a low-cost method. Dried ES sample was analyzed by X-ray diffraction, X-ray photoelectron spectroscopy and scanning electron microscopy–energy-dispersive X-ray analysis. The released amount of heavy metals increased with the increase of the content of heavy metals in ES. Electroplating enterprises will save 46.3% disposal cost by drying electroplating sludge at low-temperature.

Keywords: Water washing of heavy metals; Electroplating sludge; Hazardous waste; Low-temperature drying

1. Introduction

Hazardous waste has routinely been one of the potential issues that pollute the environment. Hazardous waste is mainly derived from the end products of many industrial and chemical activities [1]. Any release of pollutants from hazardous wastes will cause an ultimate impact on human health and potential threats to the environment [2]. Based on the newest statistics of the Chinese government, the production amount of hazardous waste was increased to 69.4 million tons in 2017 [3], most of which was generated from the raw chemical materials and chemical product industry sectors [4]. The hazardous degree of hazardous waste is related to the composition of pollutants and applied treatment methods [5]. In order to avoid the harmful effect of hazardous wastes on the environment, they must be strictly managed and reasonably treated under national and local laws [6]. In recent decades, different approaches have been attempted to dispose of hazardous waste by many researchers. The methods include harmless disposal [7], cyclic utilization [8], reduction treatment [9].

^{*} Corresponding author.

^{1944-3994/1944-3986} $\ensuremath{\mathbb{C}}$ 2021 Desalination Publications. All rights reserved.

Among these methods, cyclic utilization has been attracting more attention owing to its sustainable development, saving resources and relatively inexpensive cost [10,11]. The increasing global demand for metals makes metal recovery from continuously generated resources and waste more appealing [12]. As a result, various kinds of alternative leaching techniques, including ultrasonically enhanced two-stage acid leaching process [13], a combination of bioelectrical reactor and acid leaching method [14], and phosphate participation in the hydrochloric acid medium [15] are increasingly utilized. However, the acid pollutants generated from leaching technology demand being treated in a suitable way which needs the expenditure of a good deal of water, money and energy [16].

Harmless disposal is also commonly utilized for hazardous waste disposal due to its wide applicability [17]. The cement rotary kiln co-processing has been effectively limited for toxicity of heavy metals from hazardous waste [18]. Zhang et al. [18] found that the effect of cement rotary kiln co-processing technology is better for fixed As, Pb, Zn. Alternately, Qian et al. [19] employed MSWI fly ash-based Friedel matrices to efficaciously immobilize chromiumbearing galvanization sludge. The co-disposal of MSWI fly ash-based Friedel matrices with active aluminum is promising an available way to immobilize chromium-bearing galvanization sludge. Liu et al. [20] reported that the efficiency of cement/activated carbon (AC) solidified/stabilized (S/S) for phenol-containing hazardous wastes. The results indicated that S/S products with 2% added AC should be disposed of landfills. Therefore, those methods provide rapid curing and expand the scope of their applications and performances of solidified/stabilized for hazardous waste [21].

In order to obtain the efficiency of reduction treatment for hazardous waste, different approaches have been attempted by many researchers. Dai et al. [22] reported a direct drying method with the flue gas temperature of $100^{\circ}\text{C}-200^{\circ}\text{C}$, which removed 16%-42% PM2.5, 26%-55% PM10, and 7%-25% SO₂ from the flue gas and also preserve 95% calorific value in the sludge. Rao et al. [23] used an ultrahigh-pressure (UHP) and thin cake press dewatering device for the sludge via the addition of little or no cationic polyacrylamide and reported that the sludge with high organic matter content and thickness is hard to be dewatered.

Sludge drying could reduce the weight of sludge achieved by dehydration [24], heat treatment [25] or other methods. Numerous researches have been conducted on dehydration methods for sludge drying. The electromagnetic induction heating method was proposed by Xue et al. [26] for sludge drying. It was found that the drying rate and the release of VOCs are highly related to the working voltage. Tuncer et al. [27] numerically and experimentally investigated the experimental results of the novel convex-type solar absorber-assisted dryer and found that it shortened the drying time and promoted energy efficiency. Therefore, it is important to employ a new method for low-temperature drying of the sludge. A steam of hot dry air 50°C-70°C was utilized to pass through the sludge and direct contact between hot dry air and sludge with speed the water evaporation. Grimm et al. [28] investigated an important method that chemical sludge can be quickly dried at low-temperature. It indicated that collision between the cyclone wall and particles played a crucial role in enhancing the efficiency of heat and mass transfer. However, the release of NO_x and SO_2 increased with an increasing amount of chemical sludge in the mixtures.

The common pollutions in the exhaust gas of hazardous waste sludge include heavy metals and VOCs [25]. The direct emission of exhaust gas, which does harm to the environment, cannot be a proper solution [29]. Moreover, the cumulative cancer risks affected by the poisonous VOCs exceeded a normal risk level [30,31]. Simultaneously, electroplating sludge (ES) is a typical hazardous waste of heavy metals, a large amount of heavy metals (i.e., Cu, Cr, Ni, Zn, etc.) will be released from ES [32]. Many methods for removing heavy metals from sludge have been studied [33,34]. Tang et al. [35] investigated a biodegradable complexing agent tetrasodium of N,N-bis(carboxymethyl) glutamic acid as an electrolyte to improve heavy metals (Cu, Zn, Cr, Pb, Ni and Mn) removal efficiencies. Tang et al. [36] found rhamnolipid, saponin and sophorolipid were used to enhance heavy metals removal from the sludge in the electrokinetic tests. Other researchers also did studies in the combined rhamnolipid and saponin was used to enhance metals removal from sludge in the multiple washing steps [37]. However, there is no way to control the release of heavy metals, it will destroy the natural environment and cause potential carcinogenic risks and damage organs to human beings due to the large number of heavy metals were released from ES [38]. Therefore, searching for an appropriate method with low energy consumption and reasonable control the heavy metal release can be an important issue for researchers.

The ES drying case (Fig. S1) of the electroplating plant in Huzhou (Zhejiang Province) was investigated. It was found that the pollution of heavy metals of the exhaust gas was previously ignored in the plant. Furthermore, amounts of heavy metals of the untreated exhaust gas exceed China's National Standard and the sedimentation amount of heavy metals near the exhaust outlet also exceed the National Standard according to the detection. The amounts of heavy metals reached the National Standard after the exhaust gas was purified with water washing. Table S1 shows the comparison of the release amount of heavy metals before and after water washing, and Table S2 shows the sedimentation amount of heavy metals. Therefore, the experimental scheme to study the release effect and the control method of heavy metals in the exhaust gas of the ES drying process was studied.

In this paper, the potential environmental risks of the exhaust gas in the low-temperature drying of ES in Zhejiang were evaluated. An experimental simulation device was established. The release characteristics of heavy metals and the effective control of heavy metals during the ES dewatering process at low-temperature (50°C–70°C) were investigated. The affecting factors of temperature, the air volume and drying time on moisture content and drying rate were studied, and the release characteristics of heavy metals (i.e., Cu, Ni, Mn and Zn) of ES were summarized. Moreover, the exhaust gas was purified by the water washing in different drying conditions and the effective control of heavy metals was investigated. The influence of factors (i.e. temperature, air volume and drying time) was investigated on

the process performance to summarize a relatively optimal condition, which provides a reference for the future optimization and upgrading of low-temperature drying equipment in the field of sludge reduction.

2. Materials and methods

2.1. Agents and ES samples

In this experiment, the raw ES (stored at room temperature) was collected from a galvanization plant in Huzhou, Zhejiang. Deionized water was employed to prepare water washing. The sulfuric acid (AR, 98%) and the sodium hydroxide (AR, 99.5%) were used for the pH adjustment.

Electroplating sludge samples were collected after a plate and frame filter press in an electroplating factory in Huzhou, Zhejiang Province. A 16 mesh sieve was used to screen ES to remove the smaller particles (particle size < 1 cm). Deionized water was adjusted to the desired pH by dilute H_2SO_4 or NaOH solution to prepare water washing.

2.2. Experimental procedure

The batch experiments were performed in 250 mL wash bottles containing 70 g ES sample in the oven at a stable temperature for 1~5 h and the bottleneck was sealed with stainless steel filter. The reactor was linked with two 250 mL wash bottles containing 70.00 mL absorption solution by rubber tubes. The air volume was adjusted by a fan and a rotameter. At the end of the experiment, the ES was heated to 105°C for 5 h to obtain the solid sample and then weighted for the subsequent analysis of the dewatering rate. The laboratory setup is described in Fig. 1.

2.3. Dewatering rate

The experimental dewatering conditions (temperature, drying time, air volume) of the experiment were optimized to improve the dewatering efficiency. The moisture content and drying rate for the ES samples were calculated.

The moisture content of the ES was obtained by using the following formula:

$$\omega = \frac{G_0 - G}{G_0} \times 100\% \tag{1}$$

where ω is the moisture content of ES (%), G_0 is the initial weight of ES (g) and G is the weight of ES after drying (g).

The dewatering rate of the ES was obtained by using the formula:

$$\varphi = \frac{G_0 \omega_0 - G_1 \omega_2}{G_0 \times t} \tag{2}$$

where φ is the drying rate for ES (g g⁻¹ h⁻¹), G_0 is the initial weight of ES (g), G_1 is the dry weight of ES (g), ω_0 is the initial moisture content of ES (%), ω_1 is the moisture content of ES after drying (%), and *t* is the drying time (h).

2.4. Material characterization

The concentrations of Cu, Ni, Zn, Mn in the absorption solution were determined by the inductively coupled plasma (ICP, Thermo Fisher X Series 2). Scanning electron microscopy (SEM, ZEISS, Gemini 300) with energydispersive X-ray analysis (EDX) was used to characterize the distribution of Cu, Ni, Zn, Mn elements and surface



Fig. 1. Experimental set-up.

morphology of the solid samples. The metal species and the composition of the powder sample were obtained by X-ray photoelectron spectroscopy (XPS) analysis. The XPS analysis was carried out using the ESCALAB 250Xi with an Al K-alpha 1,486.6 eV and binding energy was calibrated with the C 1s peak (as 284.8 eV). X-ray diffraction (XRD) analysis of the dried ES powder was performed over the Bruker D8 with Cu K α -radiation at room temperature.

3. Results and discussion

3.1. Characteristics of electroplating sludge

The XRD patterns of ES samples are shown in Fig. 2. It was obvious that the characteristic peaks of $Cu(OH)_{2'}$, $Ni(OH)_{2'}$, MnO(OH), and $Zn(OH)_{2}$ were presented. The peaks at 24.01° and 33.95° (2 θ values) indicate the presence of Cu in the form of $Cu(OH)_{2'}$ peaks at 18.99°, 33.16° and 59.17° give the evidence for $Ni(OH)_{2'}$ while peaks at 19.20° and 34.00° were ascribed to that of MnO(OH) and the peaks of 31.80° and 36.29° were the characteristic peaks of Zn in the form of $Zn(OH)_{2}$. The XRD results demonstrated the existence of four heavy metal elements of Cu, Ni, Mn, Zn in ES samples.

The XPS spectra of ES are presented in Fig. 3. A typical wide scan survey spectrum of ES samples is shown in Fig S2, and the photoelectrons peaks of the survey spectrum reflect the major essentials e.g. Zn, O, Fe, Mn, Ni and C. The Cu 2p XPS spectrum displayed in Fig. 3a shows that the peak of 943.5 eV belongs to Cu^{2+} in $Cu(OH)_2$ [39]. As demonstrated in Fig. 3b, Ni 2p XPS spectrum presents photoelectron lines at the binding energy of 856.1 eV and 873.9eV, which attributed to Ni $2p_{3/2}$ and Ni $2p_{1/2}$, respectively [40]. The high-resolution XPS spectrum (Fig. 3c)

of Mn 2p is split into two sharp peaks at 641.8eV and 653.4eV, corresponding to Mn $2p_{3/2'}$ Mn $2p_{1/2}$ [41]. The high-resolution Zn 2p spectrum shows two main peaks located at 1,021.9 and 1,044.9 eV (Fig. 3d), respectively, which are attributed to the Zn $2p_{3/2}$ and Zn $2p_{1/2}$ spinorbit peaks in the spinel Zn(OH)₂ phase [42]. These results demonstrated the presence of Cu, Ni, Mn, Zn elements in the ES, which is consistent with the XRD observation.

Through XRD and XPS characterization, it was proved that ES contains heavy metals (i.e., Cu, Ni, Mn, and Zn). Furthermore, Table 1 shows the calculated content of each heavy metal in ES. Obviously, the heavy metals were the main compositions present in the dried ES sample. The ES sample contained a large amount of Zn (5.42 wt.%), and a lower amount of Cu (0.13 wt.%), Ni (0.57 wt.%), Mn (0.54 wt.%).

The micro-morphology pattern of ES was monitored by SEM as shown in Fig. 4a. The micro-morphology pattern of ES was monitored by SEM as shown in Fig. 4a. It was observed that the surface of ES basically presents a dense structure. Further, enlarge Fig. 4a and b shows that some voids are uniformly distributed on the surface of ES, which was the channels of water evaporation and heavy metal release in ES. The spatial distribution of different elements of the ES was considered by elemental mapping analysis under EDX observation (Fig. 4c–f). The elemental mapping results showed that Mn, Cu, Ni and Zn are uniformly distributed on the surface of ES samples.

3.2. Relationship between influencing factors of low-temperature drying and dehydration efficiency

The water in sludge usually existed as free water, surface water, interstitial water and bound water. Free water,



Fig. 2. XRD patterns of ES sample.



Fig. 3. (a) Cu 2p, (b) Ni 2p, (c) Mn 2p and (d) Zn 2p XPS spectra of ES sample after drying.

Table 1 Total contents of heavy metals in the ES

Element	wt.%
Water	71.6
Ni	0.57
Zn	5.42
Cu	0.13
Mn	0.54
Other	21.74

which is not bound to the particles; interstitial water, which is bound by capillary forces between the sludge floes; surface water, which is bound by adhesive forces; and intracellular water [43]. The sorting of binding energy from small to large was free water, interstitial water, surface water and bound water, which was the key factor of dehydration between water and sludge [44]. Heavy metal ions would be released with steam due to the evaporation during the dewatering of ES.

It can be seen from Fig. 5a that the moisture content of the ES sample decreased from 71.6% to 14.4%. The drying rate increased to the highest at 1 h (0.40 g g⁻¹ h⁻¹) and then steadily decreased to 0.13g g⁻¹ h⁻¹ at 5 h. According to the research elsewhere [45] and Fig. 5a, the free water was mainly evaporated at 60°C within 0–2 h in ES because the binding energy of free water was relatively low and it was easier to be broken. The moisture content of ES is 44.25% at 2 h. The interstitial water is mainly removed from 2 to 5 h due to the energy generated by temperature is enough to destroy the binding energy of interstitial water. The binding energy between surface water as well as bound water and sludge was not destroyed. Thus, the drying rate didn't drop suddenly again [45].



Fig. 4. SEM images of ES (a and b) and EDX elemental mapping of (c) Cu, (d) Ni, (e) Mn, and (f) Zn.

Fig. 5b shows that the moisture content with various air volume (0.3, 0.5, 0.7, 0.9 and 1.1 m³ h⁻¹) were 41.80%, 35.96%, 27.37%, 22.60% and 17.08%, respectively. Obviously, the drying rate gradually increased with the increase of air volume, due to the surface of sludge was dried rapidly with a large amount of air volume, which will reduce the heat conductivity of ES.

Fig. 5c shows that the moisture content gradually decreases with increasing temperature. The moisture contents at 50°C, 55°C, 60°C, 65°C and 70°C were calculated as 40.80%, 32.09%, 27.03%, 22.80% and 16.31%, respectively. The drying rate increased from 0.17 to 0.19 g g⁻¹ h⁻¹ in the range of 50°C–55°C. According to the magnitude of binding energy in ES, the free water with minimum binding energy was firstly evaporated in the range of 50°C–55°C. Moreover, the surface water with higher binding energy was mainly evaporated at higher temperature (above 55°C) in ES, and the growth of drying rate slows down.

3.3. Effect of different conditions on release amount (RA) of heavy metals from electroplating sludge under different conditions

The release amounts of heavy metals (i.e., Cu, Ni, Mn and Zn) from the ES sample are illustrated in Fig. 6. It can be observed that the release amount of heavy metals increased with the drying time, and the release amount increased steadily in the range of 2–5 h because surface water was dewatered. Simultaneously, the release amount of heavy metals increased with the air volume in the low-temperature drying process because of the accelerated evaporation of water by increased air volume. Furthermore, the release amount of Mn increased with the temperature, the release curve of Cu has a peak at 60°C, and conversely, the release amount of Ni and Zn decreased with the temperature. The reasons for this change should be considered from the sediment solubility product constant by metal ions [46] and the rest moisture content in the ES sample.



Fig. 5. Effect of (a) drying time, (b) drying temperature, and (c) air volume on moisture content and drying rate.

The sediment solubility increased with the temperature in the aspect of hydroxide precipitation. Meanwhile, the moisture content of ES decreased with the temperature, and then the reduction of moisture content further inhibited the release of heavy metals. The sediment solubility product constant of hydroxide precipitation (Cu(OH)_{2'} Ni(OH)₂, MnO(OH), and Zn(OH)₂) were 4.8×10^{-20} , 5.48×10^{-16} , 2×10^{-13} and 7.7×10^{-17} , respectively [47, 48]. As shown in Fig. 6c, the Mn release curve shows that the solubility product constant of precipitation has a greater influence than the moisture content because Mn has the maximum solubility product constant. In the range of 50° C– 70° C, the moisture content mainly affects the release of Ni and Zn,



Fig. 6. The influences of (a) drying time, (b) air volume, and (c) drying temperature on releasing the heavy metal in the ES sample.

resulting in the curve decreasing gradually. Fig. 6c shows that the released amount of Cu is dominantly influenced by solubility product constant in the range of 50°C–60°C, and the influence of moisture content on the released amount is dominant in the range of 60°C–70°C. The released amount of Zn in ES was higher than other heavy metals, because the proportion of Zn in ES was much larger than other heavy metals [49].

3.4. Influence of exhaust gas after water washing under different conditions on discharge amount (DA) of heavy metals

A series of influencing factors including drying time, air volume and drying temperature were investigated to

examine the effectiveness of heavy metals (i.e. Cu, Ni, Mn and Zn) absorption of the exhaust gas with water. The change of heavy metal contents are illustrated in Fig. 7. As shown in Fig. 7a, the heavy metal contents in the purified exhaust gas increased with the drying time, and the absorption rate of Cu in exhaust gas were 49.42%, 46.53%, 48.30%, 44.42% and 48.54% at 1, 2, 3, 4 and 5 h, respectively. The maximum absorption rates of Ni, Mn and Zn in exhaust gas at 5 h, and the maximum values were 87.32%, 93.59% and 86.19%, respectively. The minimum absorption rates were 73.32%, 83.19% and 72.93% at 1 h, respectively. The increase in temperature of the absorption solution leads to a higher absorption rate during the drying process. After water washing, the discharge amount of heavy metals all met the National Standard of China (GB 16297-1996, GB 25467-2010, GB 18485-2014).

Comparing Fig. 7b with Fig. 6b and calculating the absorption rates, it was observed that the absorption rate of Cu, Ni, Mn and Zn in exhaust gas were clearly higher at the air volume was $1.1 \text{ m}^3 \text{ h}^{-1}$. The maximum absorption rates were 60.19%, 65.15%, 87.67% and 78.04%, respectively. The absorption rate of Cu, Mn and Zn were the lowest when the air volume was 0.3 m³ h⁻¹, and the minimum values were 32.90%, 38.00% and 34.35%, respectively. The absorption rate of Ni reached a minimum of 20.47% with the air volume of 0.5 m³ h⁻¹. The excess heat of the exhaust gas led to an increase in temperature of the absorption solution, and an accelerated contact between the exhaust gas and the absorption solution with the increase of air volume, hence a better absorption efficiency of heavy metals was achieved at 1.1 m³ h⁻¹.

Fig. 7c shows that the content of Ni, Mn and Zn in exhaust gas after water washing decreased with the increase of the temperature. Dissimilarly, the content of Cu in the exhaust gas exhibited a maximum value at 50°C. The maximum absorption rates of Cu, Ni, Mn and Zn were occurred at the temperatures are 50°C, 60°C, 70°C and 65°C, respectively. Accordingly, the calculated values of absorption rate were 54.00%, 90.68%, 97.87%, 91.37%, respectively. The absorption rate of Cu decreased with the increase of temperature, indicating that Cu was sensitive to the temperature variation of the absorption solution. Meanwhile, the average absorption rates of Ni, Mn and Zn were 89.53%, 96.78% and 90.12%, respectively. Therefore, it shows that the absorption rate of Ni, Mn and Zn were relatively high and less sensitive to temperature. Hence, it is found that water washing has a practical purification effect on heavy metals in the exhaust gas under the control of three factors and the final emission contents of heavy metals conform to the National Standard in China (GB 16297-1996, GB 25467-2010, GB 18485-2014).

3.5. Effect of absorption solution with different *pH* values on heavy metals in exhaust gas

As shown in Fig. 8, the contents of Cu, Ni, Mn and Zn in exhaust gas changed after the purification by washing solution with different pH values. All the contents of Cu, Ni, Mn and Zn in the exhaust gas decreased rapidly at the first stage and then approached to a steady plateau. With the increase of pH of the absorption solution,



Fig. 7. Effect of (a) drying time, (b) air volume, and c) drying temperature affect the discharge amount of heavy metals in the exhaust gas after water washing.

the absorbed content of heavy metal ions by the solution finally approaches to stable values. The turning points showed up at the corresponding pH were 5, 3, 2 and 4 for Cu, Ni, Mn, Zn, respectively. The purification rates at the turning points of the four heavy metals reached 40.95%, 76.73%, 68.88% and 85.72%, respectively. Finally, the purification rates were 63.97%, 98.79%, 98.16% and 99.24% at pH of 13. The occurrence of a turning point in each curve was attributed to the precipitation of hydroxide corresponding to metal cations, which leads to a fixation of heavy metal



Fig. 8. The discharge amount of heavy metals in exhaust gas was purified by the solution with different pH: (a) Cu, (b) Ni, (c) Mn, and (d) Zn.

ions in the absorption solution. Accordingly, the purification effects of heavy metals (i.e., Cu, Ni, Mn and Zn) in the exhaust gas performance well at pH above 5. Therefore, it is appropriate to directly use water as a purification solution, which is consistent with the conclusion in 3.4 sections.

4. Conclusion

In this study, a control method for exhaust gas from ES reduction treatment process by low temperature drying was proposed. The results demonstrate that the drying efficiency of ES sample was the fastest at 70°C and 1.1 m³ h⁻¹, and the drying rate gradually decreases with drying time. Meanwhile, the maximum released amounts of Cu, Ni, Mn and Zn from ES reached 0.009, 0.124, 0.170 and 2.097 μ g g⁻¹, respectively, which influenced by air volume, temperature and moisture content. Batch tests showed that heavy metals in the exhaust gas had surpassed maximum national regulatory levels. Further, the absorption efficiency of heavy metals with water was obvious, especially for Ni, Mn and Zn, the absorption rate reached 90.68%, 97.87% and 91.37%, respectively. The absorption rate of Cu reached 63.67% at 60°C and 1.1 m³ h⁻¹ in this

experiment. Subsequent research that the purification effect of heavy metals in the exhaust gas increased with the increase pH of absorption solution. The purification rates for Cu, Ni, Mn and Zn were 63.97%, 98.79%, 98.16% and 99.24%, respectively when pH was 13. The exhaust gas water washing test indicated that the risk of releasing heavy metals from ES would be reduced substantially and satisfied the regulatory standard. Therefore, knowledge of the release mechanism of heavy metals from ES is useful for determining the economical control method for efficient and effective absorb of heavy metal elements during the low temperature drying process. Electroplating sludge dried at low temperature, because of the weight reduction, its disposal cost can be reduced by 46.3%, the specific data were shown in Table S3. However, the main disadvantage of low-temperature drying method was the long time of the process.

Acknowledgements

All authors contributed to the study's conception and design. Material preparation, data collection and analysis were performed by Xiaoting Hong and Huaiyu Liu. The first draft of the manuscript was written by Huaiyu Liu and all authors commented on previous versions of the manuscript. Kwan San Hui, Miao Deng, Keming Wu, and Yingying Du contribute to the revision of this article. All authors read and approved the final manuscript.

Formatting of funding sources

Financial supports for this work were provided by the Natural Science Foundation of Zhejiang Province (LY18E080016), the Open Foundation of State Environmental Protection Key Laboratory of Mineral Metallurgical Resources Utilization and Pollution Control (HB201909), and the Fundamental Research Funds of Zhejiang Sci-Tech University (2020Q048).

References

- [1] T.K. Cherniaeva, Actual problems of the impact of production and management of industrial waste on the environment and public health (review of literature), Gig. Sanit., (2013) 32–35.
- [2] M. Triassi, R. Alfano, M. Illario, A. Nardone, O. Caporale, P. Montuori, Environmental pollution from illegal waste disposal and health effects: a review on the "triangle of death", Int. J. Environ. Res. Public Health, 12 (2015) 1216–1236.
- [3] R.J. Slack, J. Gronow, N. Voulvoulis, Household hazardous waste in municipal landfills: contaminants in leachate, Sci. Total Environ., 337 (2005) 119–137.
- [4] H.B. Duan, Q.F. Huang, Q. Wang, B.Y. Zhou, J.H. Li, Hazardous waste generation and management in China: a review, J. Hazard. Mater., 158 (2008) 221–227.
- [5] A.A. Namen, C. Brasil Fda, J.J. Abrunhosa, G.G. Abrunhosa, R.M. Tarre, F.J. Marques, RFID technology for hazardous waste management and tracking, Waste Manage. Res., 32 (2014) 59–66.
- [6] N.T.K. Thai, Hazardous industrial waste management in Vietnam: current status and future direction, J. Mater. Cycles Waste Manage., 11 (2009) 258–262.
- [7] S. Oh, R. Bade, F. Li, W.S. Shin, Solidification/stabilization of heavy metals in tannery sludge char with various binders, Desal. Water Treat., 52 (2014) 889–899.
- [8] Z. Phua, A. Giannis, Z.L. Dong, G. Lisak, W.J. Ng, Characteristics of incineration ash for sustainable treatment and reutilization, Environ. Sci. Pollut. Res., 26 (2019) 16974–16997.
- [9] A. An-nori, L. El Fels, A. Ezzariai, M. El Gharous, K. El Mejahed, M. Hafidi, Effects of solar drying on heavy metals availability and phytotoxicity in municipal sewage sludge under semiarid climate, Environ. Technol. Innovation, 19 (2020) 101039, doi: 10.1016/j.eti.2020.101039.
- [10] N. Chandra, S.S. Amritphale, D. Pal, Manganese recovery from secondary resources: a green process for carbothermal reduction and leaching of manganese bearing hazardous waste, J. Hazard. Mater., 186 (2011) 293–299.
- [11] V. Gunarathne, A.U. Rajapaksha, M. Vithanage, N. Adassooriya, A. Cooray, S. Liyanage, B. Athapattu, N. Rajakaruna, A.D. Igalavithana, D. Hou, D.S. Alessi, Y.S. Ok, Heavy metal dissolution mechanisms from electrical industrial sludge, Sci. Total Environ., 696 (2019) 133922, doi: 10.1016/j. scitotenv.2019.133922.
- [12] K. Chandrasekhar, G. Kumar, S.V. Mohan, A. Pandey, B.-H. Jeon, M. Jang, S.H. Kim, Microbial Electro-Remediation (MER) of hazardous waste in aid of sustainable energy generation and resource recovery, Environ. Technol. Innovation, 19 (2020) 100997, doi: 10.1016/j.eti.2020.100997.
- [13] C.C. Li, F.C. Xie, Y. Ma, T.T. Cai, H.Y. Li, Z.Y. Huang, G.Q. Yuan, Multiple heavy metals extraction and recovery from hazardous electroplating sludge waste via ultrasonically enhanced twostage acid leaching, J. Hazard. Mater., 178 (2010) 823–833.
- [14] P. Wu, L.-j. Zhang, C.-b. Lin, X.-x. Xie, X.-y. Yong, X.-y. Wu, J. Zhou, H.-h. Jia, P. Wei, Extracting heavy metals from

electroplating sludge by acid and bioelectrical leaching using *Acidithiobacillus ferrooxidans,* Hydrometallurgy, 191 (2020) 105225, doi: 10.1016/j.hydromet.2019.105225.

- [15] R. Su, B. Liang, J. Guan, Leaching effects of metal from electroplating sludge under phosphate participation in hydrochloric acid medium, Procedia Environ. Sci., 31 (2016) 361–365.
- [16] R. Golmohammadzadeh, F. Faraji, F. Rashchi, Recovery of lithium and cobalt from spent lithium ion batteries (LIBs) using organic acids as leaching reagents: a review, Resour. Conserv. Recycl., 136 (2018) 418–435.
- [17] F. Galliou, N. Markakis, M.S. Fountoulakis, N. Nikolaidis, T. Manios, Production of organic fertilizer from olive mill wastewater by combining solar greenhouse drying and composting, Waste Manage., 75 (2018) 305–311.
- [18] J.-l. Zhang, J.-g. Liu, C. Li, Y.-y. Jin, Y.-f. Nie, Comparison of fixation effects of heavy metals between cement rotary kiln co-processing and cement solidification/stabilization, Huan jing ke xue, 29 (2008) 1138–1142.
- [19] G. Qian, X. Yang, S. Dong, J. Zhou, Y. Sun, Y. Xu, Q. Liu, Stabilization of chromium-bearing electroplating sludge with MSWI fly ash-based Friedel matrices, J. Hazard. Mater., 165 (2009) 955–960.
- [20] J.G. Liu, X.Q. Nie, X.W. Zeng, Z.J. Su, Long-term leaching behavior of phenol in cement/activated-carbon solidified/ stabilized hazardous waste, J. Environ. Manage., 115 (2013) 265–269.
- [21] D.S. Vijayan, D. Parthiban, Effect of solid waste based stabilizing material for strengthening of expansive soil – a review, Environ. Technol. Innovation, 20 (2020) 101108, doi: 10.1016/j. eti.2020.101108.
- [22] Z. Dai, M. Su, X. Ma, G. Wang, D. Li, C. Liu, H. Weng, Direct thermal drying of sludge using flue gas and its environmental benefits, Drying Technol.: An Int. J., 36 (2018) 1006–1016.
- [23] B. Rao, Y. Zhu, M. Yu, X. Lu, Y. Wan, G. Huang, X. Su, X. Liu, High-dry dewatering of sludge based on different pretreatment conditions, Process Saf. Environ. Prot., 122 (2019) 288–297.
- [24] H. Liu, H. Xiao, B. Fu, H. Liu, Feasibility of sludge deepdewatering with sawdust conditioning for incineration disposal without energy input, Chem. Eng. J., 313 (2017) 655–662.
- [25] Z.-x. Tan, J.-h. Yan, X.-g. Jiang, H.-d. Xue, Y. Chi, Thermal analysis and the distribution rule of heavy metals during electroplating sludge combustion, Huan jing ke xue, 27 (2006) 998–1002.
- [26] Y. Xue, C. Wang, Z. Hu, Y. Zhou, G. Liu, H. Hou, Y. Xiao, T. Wang, J. Li, Thermal treatment on sewage sludge by electromagnetic induction heating: methodology and drying characterization, Waste Manage., 78 (2018) 917–928.
- [27] A.D. Tuncer, A. Sözen, F. Afshari, A. Khanlari, C. Şirin, A. Gungor, Testing of a novel convex-type solar absorber drying chamber in dehumidification process of municipal sewage sludge, J. Cleaner Prod., 272 (2020).
- [28] A. Grimm, J. Etula, R. Salh, G. Kalén, M. Segerström, J. Brücher, C. Söderberg, D. Soukup, C. Pfeifer, S.H. Larsson, Slagging and fouling characteristics during co-combustion of Scots pine bark with low-temperature dried pulp and paper mill chemical sludge, Fuel Process. Technol., 193 (2019) 282–294.
- [29] J. Latosinska, Risk assessment of soil contamination with heavy metals from sewage sludge and ash after its incineration, Desal. Water Treat., 199 (2020) 297–306.
- [30] S.A. Khan, S. Muhammad, S. Nazir, F.A. Shah, Heavy metals bounded to particulate matter in the residential and industrial sites of Islamabad, Pakistan: implications for non-cancer and cancer risks, Environ. Technol. Innovation, 19 (2020) 100822, doi: 10.1016/j.eti.2020.100822.
- [31] M.J. Kim, Y.K. Seo, J.H. Kim, S.O. Baek, Impact of industrial activities on atmospheric volatile organic compounds in Sihwa-Banwol, the largest industrial area in South Korea, Environ. Sci. Pollut. Res., 27 (2020) 28912–28930.
- [32] N. Sezgin, N. Balkaya, Removal of heavy metal ions from electroplating wastewater, Desal. Water Treat., 93 (2017) 257–266.

- [33] J. Tang, J. He, H. Tang, H. Wang, W. Sima, C. Liang, Z. Qiu, Heavy metal removal effectiveness, flow direction and speciation variations in the sludge during the biosurfactantenhanced electrokinetic remediation, Sep. Purif. Technol., 246 (2020), doi: 10.1016/j.seppur.2020.116918.
- [34] J. Tang, Z. Qiu, H. Tang, H. Wang, W. Sima, C. Liang, Y. Liao, Z. Li, S. Wan, J. Dong, Coupled with EDDS and approaching anode technique enhanced electrokinetic remediation removal heavy metal from sludge, Environ. Pollut., 272 (2021) 115975, doi: 10.1016/j.envpol.2020.115975.
- [35] J. Tang, J. He, T. Liu, X. Xin, H. Hu, Removal of heavy metal from sludge by the combined application of a biodegradable biosurfactant and complexing agent in enhanced electrokinetic treatment, Chemosphere, 189 (2017), doi: 10.1016/j. chemosphere.2017.09.104.
- [36] J. Tang, J. He, X. Xin, H. Hu, T. Liu, Biosurfactants enhanced heavy metals removal from sludge in the electrokinetic treatment, Chem. Eng. J., 334 (2018), doi: 10.1016/j.cej.2017.12.010.
 [37] J. Tang, J. He, Z. Qiu, X. Xin, Metal removal effectiveness,
- [37] J. Tang, J. He, Z. Qiu, X. Xin, Metal removal effectiveness, fractions, and binding intensity in the sludge during the multiple washing steps using the combined rhamnolipid and saponin, J. Soils Sediments, 19 (2019) 1286–1296.
- [38] Q. Zhang, L. Zhang, W.J. Sang, M. Li, W.H. Cheng, Chemical speciation of heavy metals in excess sludge treatment by thermal hydrolysis and anaerobic digestion process, Desal. Water Treat., 57 (2016) 12770–12776.
- [39] W. Shi, J. Lian, Fluffy intersected NiCo–OH nanosheet decorated hollow Cu(OH), nanotube arrays on Cu foam for high-performance Ni–Zn battery, J. Solid State Chem., 290 (2020) 121569, doi: 10.1016/j.jssc.2020.121569.
- [40] S. Zhou, C. Hao, J. Wang, X. Wang, H. Gao, Metal-organic framework templated synthesis of porous NiCo₂O₄/ZnCo₂O₄/ Co₃O₄ hollow polyhedral nanocages and their enhanced pseudocapacitive properties, Chem. Eng. J., 351 (2018) 74–84.

- [41] W.S. Shin, K.R. Na, Y.K. Kim, Adsorption of metal ions from aqueous solution by recycled aggregate: estimation of pretreatment effect, Desal. Water Treat., 57 (2016) 9366–9374.
- [42] S.U. Awan, S.K. Hasanain, J. Rashid, S. Hussain, S.A. Shah, M.Z. Hussain, M. Rafique, M. Aftab, R. Khan, Structural, optical, electronic and magnetic properties of multiphase ZnO/ Zn(OH)₂/ZnO₂ nanocomposites and hexagonal prism shaped ZnO nanoparticles synthesized by pulse laser ablation in heptanes, Mater. Chem. Phys., 211 (2018) 510–521.
- [43] P. Lowe, Developments in the thermal drying of sewage sludge, Water Environ. J., 9 (1995) 306–316.
- [44] W. Yu, Y. Wan, Y. Wang, Y. Zhu, S. Tao, Q. Xu, K. Xiao, S. Liang, B. Liu, H. Hou, J. Hu, J. Yang, Enhancing waste activated sludge dewaterability by reducing interaction energy of sludge flocs, Environ. Res., 196 (2020) 110328, doi: 10.1016/j. envres.2020.110328.
- [45] G. Chen, P. Lock Yue, A.S. Mujumdar, Sludge dewatering and drying, Drying Technol., 20 (2002) 883–916.
- [46] M. Toribio, J. Romanyà, Leaching of heavy metals (Cu, Ni and Zn) and organic matter after sewage sludge application to Mediterranean forest soils, Sci. Total Environ., 363 (2006) 11–21.
- [47] S. Haas, V. Boschi, A. Grannas, Metal sorption studies biased by filtration of insoluble metal oxides and hydroxides, Sci. Total Environ., 646 (2019) 1433–1439.
- [48] G. Gnanaprakash, J. Philip, B. Raj, Effect of divalent metal hydroxide solubility product on the size of ferrite nanoparticles, Mater. Lett., 61 (2007) 4545–4548.
- [49] T. Wang, S. Huang, T. Fu, J. Li, X. Zhou, Y. Xue, H. Hou, Effects of electromagnetic induction on migration and speciation of heavy metals in drying sewage sludge: mechanistic insights, Waste Manage., 109 (2020) 192–201.

86

Supporting information

Table S1

Release amount of heavy metals before and after water washing at electroplating factory in Huzhou

	Cr	Ni	Cu	Zn
#1 (µg/m ³)	258	3,990	286	8,595
#2 (µg/m³)	2.9	9.76	9.24	43.2
National Standard values (µg/m³)	<7.5	<40	<60	<500
Remarks	GB 16297-1996	GB 25467-2010	GB 18485-2014	GB 16297-1996

Note: #1 the amounts of heavy metals of the unpurified exhaust gas exceed China's National Standard;

#2 the amounts of heavy metals of the purified exhaust gas met China's National Standard.

Table S2

Sedimentation amount of heavy metals at electroplating factory in Huzhou.

	Cr	Ni	Cu	Zn
#1 (μg/m³)	158	2863	186	6754
#2 (µg/m³)	1.2	5.6	3.8	32.8

Note: #1 the sedimentation amounts of heavy metals of the unpurified exhaust gas;

#2 the sedimentation amounts of heavy metals of the purified exhaust gas.

Table S3

Calculation of electroplating sludge disposal cost

Item	Moisture content (%)	Weight (t/d)	Disposal cost (RMN/t)	Drying cost (RMB/t)	Disposal cost (RMB/d)
Untreated electroplating sludge	71.6	5	1,500	0	7,500
Drying treatment of electroplating sludge	20	1.78	1,700	200	4,026
Cost saving	-	-	-	-	3,474

Note: The drying cost was calculated by the amount of untreated electroplating sludge; Electroplating enterprises will save 46.3% disposal cost.