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Characteristics of solid waste from common generation source in nonferrous smelting industry reveal a new classification method

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ABSTRACT

Solid waste produced by the nonferrous smelting industry has a significant number of notable differences. The lack of recognition of solid waste characteristics is the main factor restricting its disposal and utilization. In this study, we analyzed the main production processes of the nonferrous smelting industry; identified the key production nodes of solid waste; and clarified the characteristics, including the physical, chemical, and pollution characteristics of solid wastes, through a large sample statistical analysis. We found similarities among solid wastes from a common generation source as well as notable differences among the different generation sources: slags and sludges from waste acid treatment and wastewater treatment units had a water content of 27.43–52.71% and 51.14–68.27%, respectively, which were significantly higher than those of other metallurgy and dust collection units; the pH of slags from an electrorefining unit was strongly alkaline; the mineral phase of sludges from wastewater treatment was only calcite; slags from a waste acid treatment unit were mainly in phase of gypsum, claudetite, and anglesite; the chemical composition of slags from pyrometallurgy and hydrometallurgy units was mainly SiO_2 and Fe_2O_3. In this paper, we discuss a new classification method based on a common generation source for the first time. These results are beneficial to guide the disposal, utilization, and management of solid waste.

1. Introduction

With complex processes and many production nodes, solid waste produced by the nonferrous smelting industry has a significant number of notable differences [1,2]. Their complex composition and heavy metal toxic substances may cause environmental risks during disposal or utilization process [3,4]. The ability to clarify the characteristics of these solid wastes holds great significance for guiding waste disposal and utilization [5]. Illegal dumping of solid waste has occurred frequently in recent years, the lack of recognition of solid waste characteristics in the nonferrous smelting industry, however, has become the main factor restricting the

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identification and management of solid wastes of unknown origin [6–8].

In this study, we found that the characteristics of solid waste were closely related to the generation source, and the composition characteristics of solid waste from the common generation source were relatively similar. For example, the sludge produced in the solid–liquid separation process of sewage treatment was mostly gypsum and calcite in terms of phase [9], and the dust produced in the gas–solid separation process of waste gas treatment was mostly powder and particle in terms of apparent morphology [10]. Therefore, a classification method based on the common generation source will benefit the disposal and utilization of solid waste.

For the difference of industrial structures between different countries, the classification methods and management mechanisms of solid waste are also quite different. According to the Resource Conservation and Regeneration Act (RCRA) [11], solid waste in the United States is divided into general solid waste and hazardous waste [12], and hazardous wastes are classified according to their hazardous characteristics and sources. The European Waste List classifies solid waste into 20 categories based on a combination of industry sources and types of waste [13]. In Russia, the solid waste classification method is constructed primarily according to the hazardous characteristics by the Russian Waste Classification List [14].

The hazardous waste classification method in China is based primarily on the National Hazardous Waste List [15], which divides the source and category of hazardous waste in the mainstream production process of major industrial sectors. In this way, a variety of solid waste, such as dust and sludge from the same industry with different composition characteristics, may be classified into one category. Because of the different generation sources of dust and sludge, the composition characteristics of the two solid wastes are quite different. Moreover, among the different categories of hazardous waste, other waste categories have similar utilization and disposal characteristics. For example, 101 types of wastewater treatment sludge are distributed in 30 categories in the list.

In fact, all of the existing solid waste classification methods ignore the principle that common generation source waste has similar characteristics and that these methods do not use the composition and pollution characteristics of the waste [16,17]. In our study, we analyzed the main production processes of the nonferrous smelting industry, identified the key production nodes of solid waste, clarified the waste characteristics from different generating units, and found a solid waste classification method based on the common generation source. These results could be used to guide the disposal and utilization of solid wastes from the nonferrous smelting industry, and may lay the foundation to establish a new industrial solid waste classification system in the future, which would be useful to identify and source tracing of industrial solid wastes of unknown origin.

2. Materials and methods

We collected 30 solid waste samples from six copper (Cu), lead (Pb), zinc (Zn), and aluminum (Al) smelting plants in Henan, Chongqing, and Shandong. We followed sampling methods in accordance with the *Technical Specifications on Sampling and Sample Preparation from Industry Solid Waste (HJ/T 20–1998).* [18] Sample names and production processes are presented in Table 1.

Table 1
Sample names and production processes.

Sample Number	Samples	Production processes
1	Smelting slag	Pyrometallurgy of copper
2	Blowing slag	Pyrometallurgy of copper
3	Water quenched slag	Pyrometallurgy of lead
4	Volatile kiln slag	Hydrometallurgy of zinc
5	Purifying slag	Hydrometallurgy of zinc
6	Roasting and leaching slag	Hydrometallurgy of zinc
7	Zinc scum	Hydrometallurgy of zinc
8	Copper removal slag	Pyrometallurgy of lead
9	Refining slag	Pyrometallurgy of lead
10	Overhaul slag	Electrolysis of aluminum
11	Aluminum ash slag	Electrolysis of aluminum
12	Secondary aluminum dross	Electrolysis of aluminum
13	Salt slag	Electrolysis of aluminum
14	Smelting dust	Pyrometallurgy of copper
15	Blowing dust	Pyrometallurgy of copper
16	Reduction furnace dust	Pyrometallurgy of lead
17	Fume furnace dust	Pyrometallurgy of lead
18	Roasting dust	Hydrometallurgy of zinc
19	Calcinating dust	Secondary aluminum metallurgy
20	Casting furnace dust	Electrolysis of aluminum
21	Recycled aluminum smelting dust	Secondary aluminum metallurgy
22	Refined dust	Secondary aluminum metallurgy
23	Arsenic slag	Pyrometallurgy of copper
24	Copper neutralizing slag	Pyrometallurgy of copper
25	Acid mud	Pyrometallurgy of copper
26	Zinc neutralizing slag	Hydrometallurgy of zinc
27	Gypsum slag	Pyrometallurgy of copper
28	Lead neutralizing slag	Pyrometallurgy of lead
29	Lead water treated sludge	Pyrometallurgy of lead
30	Zinc water treated sludge	Hydrometallurgy of zinc

We detected chemical composition according to the *General Rules for Wavelength Dispersive X–ray Fluorescence Spectrometry (JY/T 0569–2020)* [19]. We detected the mineral phase according to the *General Rules for X–ray Polycrystalline Diffractometry (JY/T 009–1996)* [20]. We detected the heavy metal concentration according to the *Identification Standards for Hazardous Wastes—Identification for Toxic Substance Content (GB 5085.6–2007)* [21]. We detected the leaching toxicity concentration according to the *Identification Standard for Hazardous Wastes—Identification for Extraction Toxicity (GB 5085.3–2007)* [22]. Quality control and quality assurance were conducted in strict accordance with the corresponding standards.

3. Results and discussions

3.1. Classification method of common generation sources

Nonferrous smelting is a production activity that uses pyrometallurgy, hydrometallurgy, or chemical methods to extract metals from ores, reduce impurities contained in metals or increase certain components of metals, and then produce required metals. The solid wastes of nonferrous smelting in our study came from the process of Cu, Pb, Zn, and Al by pyrometallurgy, hydrometallurgy, and secondary metallurgy.

Flash smelting with flash blowing and continuous Cu smelting with double-bottom blowing are commonly used in Cu pyrometallurgy [23]. Cu concentrate, including pyrite and chalcopyrite with flux and Cu–bearing ingredients, was dried and sent to a smelting furnace for Cu matte production, and then crude Cu was produced by blowing furnace. The crude Cu was refined by an anode furnace and cast into an anode plate, which was electrolytically refined to produce cathode Cu with a purity of 99.99%. The main smelting section produced smelting slag, blowing slag, refining slag, and other solid waste, and dusts were produced from the smelting furnace, blowing furnace, and anode furnace flue gas dust collection process. Anode scrap and anode mud were produced from electrolytic refining process, and acid mud and neutralizing slag were produced from the waste acid treatment process.

Lead metallurgy refers to the production of Pb products with Pb concentrate or Pb waste as raw materials [24–26]. Lead pyrometallurgy includes oxygen–rich smelting with direct reduction and oxygen–rich flash smelting, which mainly consists of smelting of Pb concentrate, refining of crude Pb, and recovery of precious metals. Lead slag, high-calcium slag, and waste acid treatment sludge are produced mainly from waste acid treatment process, recovery slag is produced mainly from precious metal recovery process, and water quenching slag is produced mainly from the fuming furnace slag water quenching process.

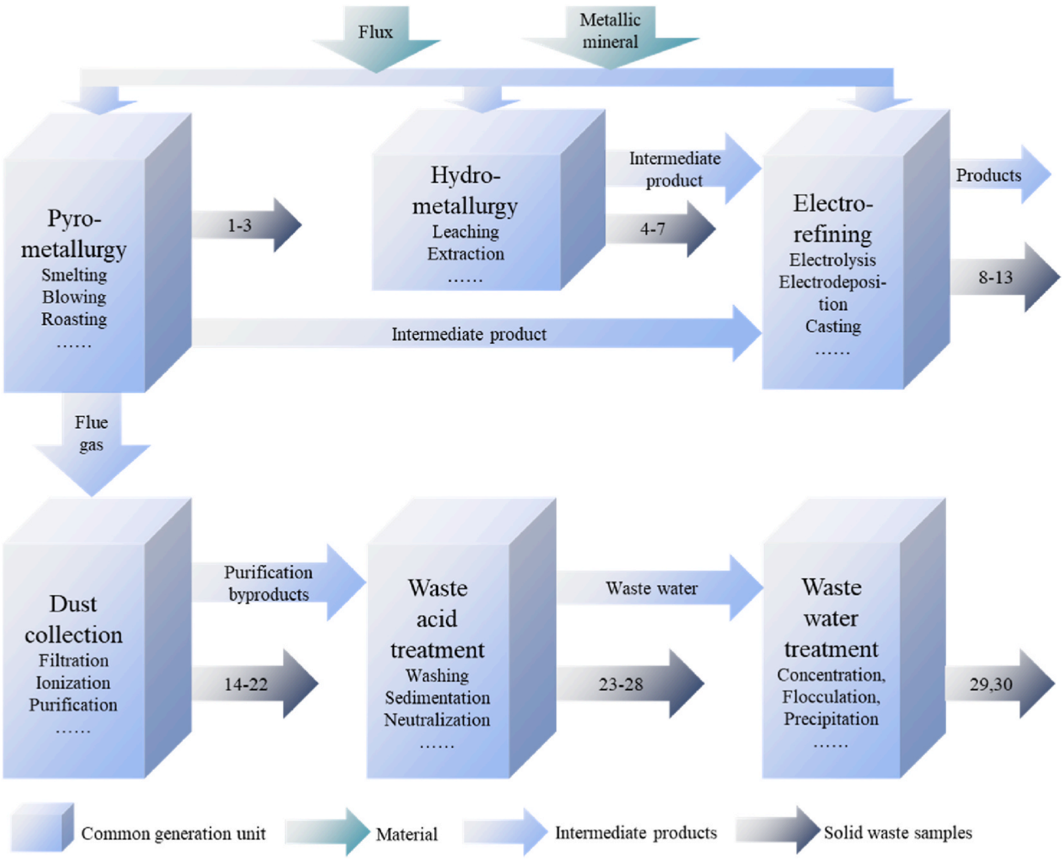

Fig. 1. Common generation units and production process.

Zinc metallurgy refers to the production of Zn products with Zn concentrate or Zn-containing waste as raw materials, mainly hydrometallurgy [27,28]. Zinc concentrate or Zn–bearing material can be added to a roaster, through air- or oxygen-rich roasting. The reaction of Zn sulfide into Zn oxide and Zn sulfate occurs under high temperature, then through a neutral and acidic solution for leaching and purification, and finally produces Zn products through electrolytic refining. Zinc hydrometallurgy includes roasting, leaching, purification, and electrodeposition. Acid mud is produced in the dust collection and purification process of flue gas, kiln slag is produced in the evaporation kiln treatment process after leaching hydraulic filtration, and Pb mud is produced in the leaching process after roasting Zn oxide.

Aluminum metallurgy refers to the production of refining Al products by smelting, electrolysis, casting, or melting of bauxite or waste Al [29,30]. It includes alumina, electrolytic Al, and secondary Al metallurgy, with alumina production primarily following the Bayer method. The two major processes include the sodium–aluminate solution crystal–seed decomposition process and the use of seed mother liquor leaching bauxite. The production of electrolytic Al uses large prebaking cell technology. A direct current flows into the electrolytic cell, and an electrochemical reaction occurs on the cathode and anode. Aluminum dross, overhaul slag, aluminum ash slag, anode scrap, and secondary aluminum dross are produced from the electrolytic refining process, and the dust collected from the melting casting furnace is produced from the Al ingot process.

According to this metallurgy process principle analysis, we categorized the main production processes of the nonferrous smelting industry into six generating units: pyrometallurgy (U1), hydrometallurgy (U2), electrorefining (U3), dust collection (U4), waste acid treatment (U5), and wastewater treatment (U6). Pyrometallurgy includes smelting, blowing, roasting, and other reaction processes; hydrometallurgy includes leaching, extraction, and other reaction processes; electrorefining includes electrolysis, electrodeposition, melting, and casting and other reaction processes; dust collection includes filtration, ionization, purification, and other processes; waste acid treatment includes washing, sedimentation, neutralization, and other reaction processes; and wastewater treatment includes concentration, flocculation, precipitation, and other reaction processes. The main production process and common generation nodes of solid waste from the nonferrous smelting industry are shown in Fig. 1.

3.2. Physical characteristics of solid waste from common generation source

The water content and pH of solid waste from six common generation units are shown in Fig. 2. Slags from pyrometallurgy, hydrometallurgy, and electrorefining units had a water content of 0.02–1.12%, 1.93–25.13%, and 0.17–3.87%, respectively. Dust from the dust collection unit had a water content of 0.27–2.59%. Slags and sludges from the waste acid treatment and wastewater treatment units had a water content of 27.43–52.71% and 51.14–68.27%, respectively, which were significantly higher than those from the other generating units.

The pH of solid wastes from six generation units was in the range of 6.43–8.92, 3.57–9.48, 10.86–13.41, 1.85–11.48, 0.61–13.67, and 9.02–9.21, respectively. Slags from pyrometallurgy and hydrometallurgy units were neutral or had weak acidity and weak alkalinity. For slags from electrorefining units, however, the pH was strongly alkaline because during electrolysis, the anode was alkaline and impurities precipitated out at the anode [31]. Dusts and sludges from dust collection and wastewater treatment units were mainly neutral. For slags from waste acid treatment, in which arsenic slag and acid mud were strongly acidic, neutralizing slag and gypsum slag were strongly alkaline. We found that the production process determined the acidic or alkaline properties of the solid wastes. The sulfur dioxide flue gas produced in the smelting process was washed by acid in the purification procedure with a strongly acidic environment, and acid mud was produced after precipitation. In contrast, sodium hydride sulfide was added to As containing waste acid produced during the purification procedure to remove As impurities, and then As was produced [32]. After removing the As, the waste acid entered a neutralization procedure by adding carbide slag, which was composed of calcium hydroxide with strong

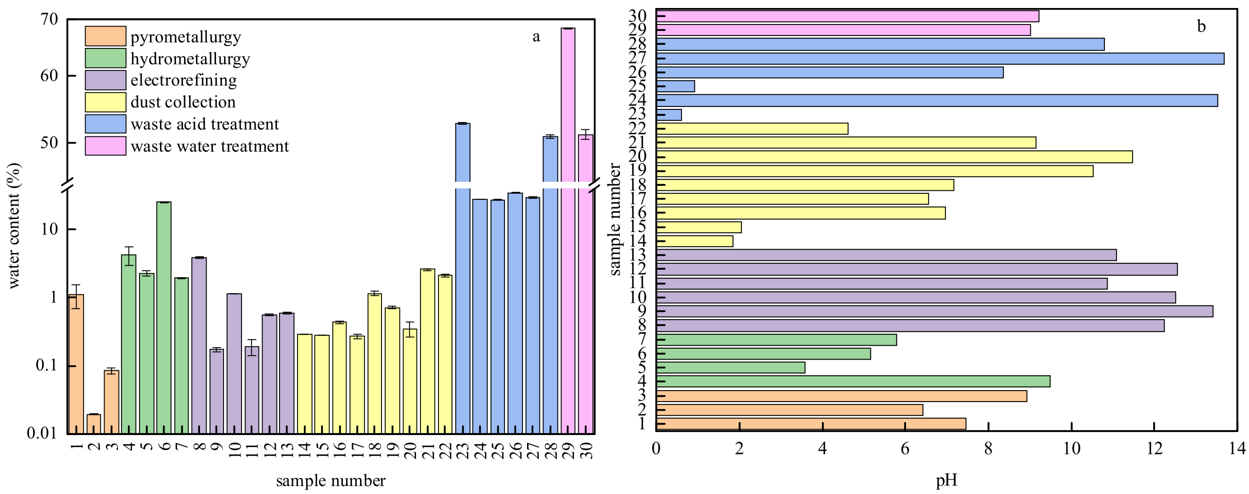

Fig. 2. Water content (a) and pH (b) of solid waste from six common generation units.

alkalinity. Then, neutralizing slag and gypsum slag were produced [33].

3.3. Chemical characteristics of solid waste from common generation source

The mineral phase and chemical composition of solid waste from six common generation units are shown in Figs. 3 and 4. Slags from the pyrometallurgy unit were in the phase of fayalite ($2FeO \cdot SiO_2$), magnetite (FeO, Fe_2O_3, Fe_3O_4), hedenbergite ($CaFeSi_2O_6$), and chalcocite ($Cu_{1.96}S$), and the water-quenched slag was amorphous. Slags from the hydrometallurgy unit were in the phase of magnetite (FeO, Fe_2O_3, Fe_3O_4), hedenbergite ($CaFeSi_2O_6$), domeykite (Cu_3As), and zincite (ZnO). Meanwhile, the chemical composition of slags from these two units was mainly SiO_2 and Fe_2O_3, which corresponded to the mineral phase. The content of SiO_2 was 10.01–33.69% and 0.546–7.14% in these two units, and Fe_2O_3 had a content of 18.71–50.81% and 0.272–25.42%, respectively.

In the electrorefining unit, slags were in the phase of claudetite (As_2O_3), massicot (PbO), cryolite (Na_3AlF_6), corundum (Al_2O_3), and spinel ($MgAl_2O_4$). Na_3AlF_6 was the main raw material for electrolysis of Al. Dusts from the dust collection unit were in the phase of zincite (ZnO), anglesite ($PbSO_4$), claudetite (As_2O_3), periclase (MgO), and chalcocyanite ($CuFeS_2$), and they usually were similar to the raw ore in terms of mineral phase. The chemical composition was mainly Al_2O_3, which had a content of 37.06–83.98% in the electrorefining unit and 0.08–69.57% in the dust collection unit. Samples in our study from these two generation units were mostly from the Al-smelting process.

Slags from the waste acid treatment unit were in the phase of gypsum ($CaSO_4 \cdot 2H_2O$), claudetite (As_2O_3), and anglesite ($PbSO_4$). In this unit, neutralizing slag and gypsum slag were produced using the neutralization procedure by adding calcium hydroxide [33], which made the main phase of $CaSO_4 \cdot 2H_2O$. The mineral phase of sludges from wasterwater treatment was only calcite ($CaCO_3$). Sodium carbonate usually is used to remove calcium ions from waste water, which accounted for the main phase of $CaCO_3$ in sludges. The chemical composition was CaO, which had a content of 0.2–45.8% and 18.5–27.22% in these two units.

3.4. Pollution characteristics of solid waste from common generation source

The heavy metal concentration and leaching toxicity of solid waste from six common generation units are shown in Fig. 5. Cu was the heavy metal with the highest concentration in slags from the pyrometallurgy unit, which ranged from 19840.8 to 347230 mg/kg. Slags from the hydrometallurgy unit had a high concentration of Cr, which ranged from 5081.08 to 151343 mg/kg. The concentration of Cu ranged from 475.19 to 106544 mg/kg and that of Zn ranged from 10920.6 to 190054 mg/kg in slags from the hydrometallurgy unit. In the electrorefining units, slags had a high concentration of Cu, which ranged from 624.79 to 69538 mg/kg, and the concentration of Pb ranged from 34.31 to 50395 mg/kg. In the dust collection unit, dusts had a high concentration of Zn, which ranged from 520.24 to 87137.6 mg/kg, and the concentration of Cu ranged from 82.15 to 99136.8 mg/kg. In the waste acid treatment unit, slags had a high concentration of Cr, which ranged from 979.72 to 98038.3 mg/kg, and the concentration of As ranged from 129.6 to

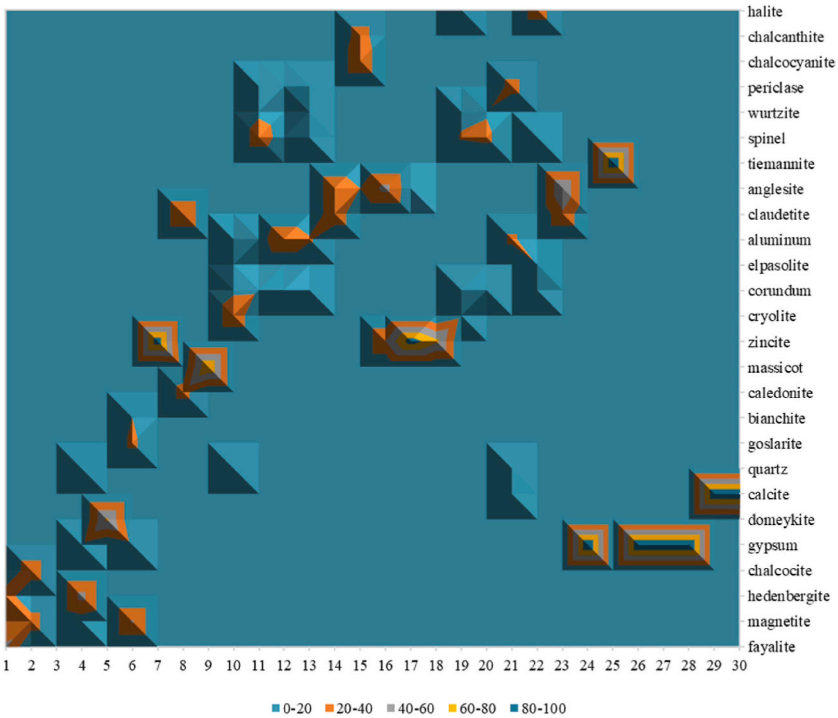

Fig. 3. Mineral phase of solid waste from six common generation units.

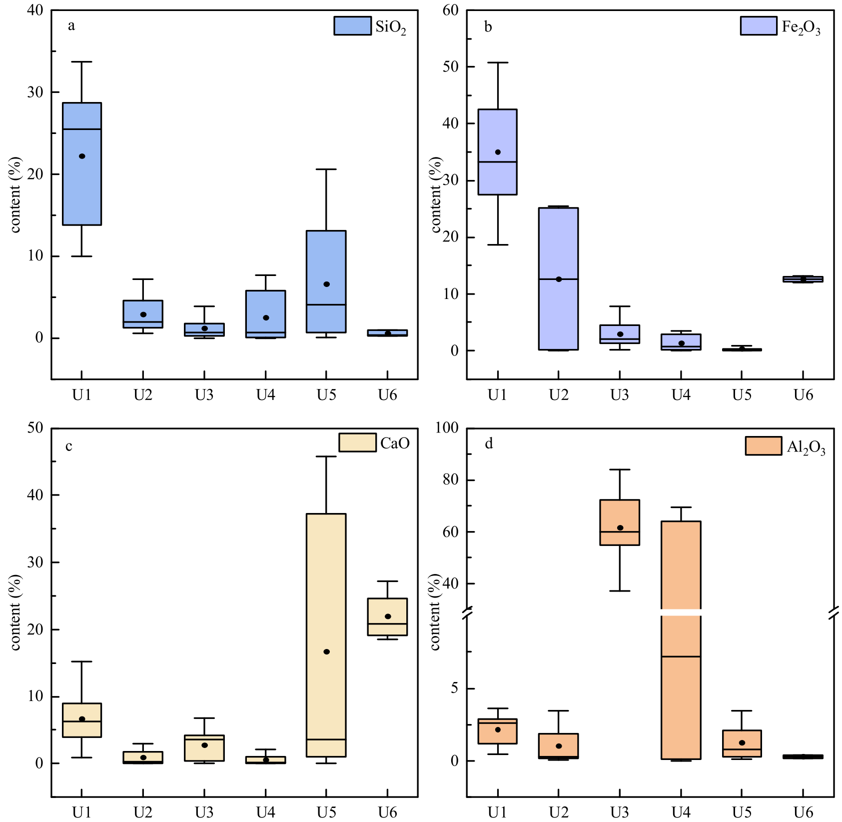

Fig. 4. Chemical composition of solid waste from six common generation units, figure a, b, c, and d show content of SiO_2, Fe_2O_3, CaO and Al_2O_3, respectively.

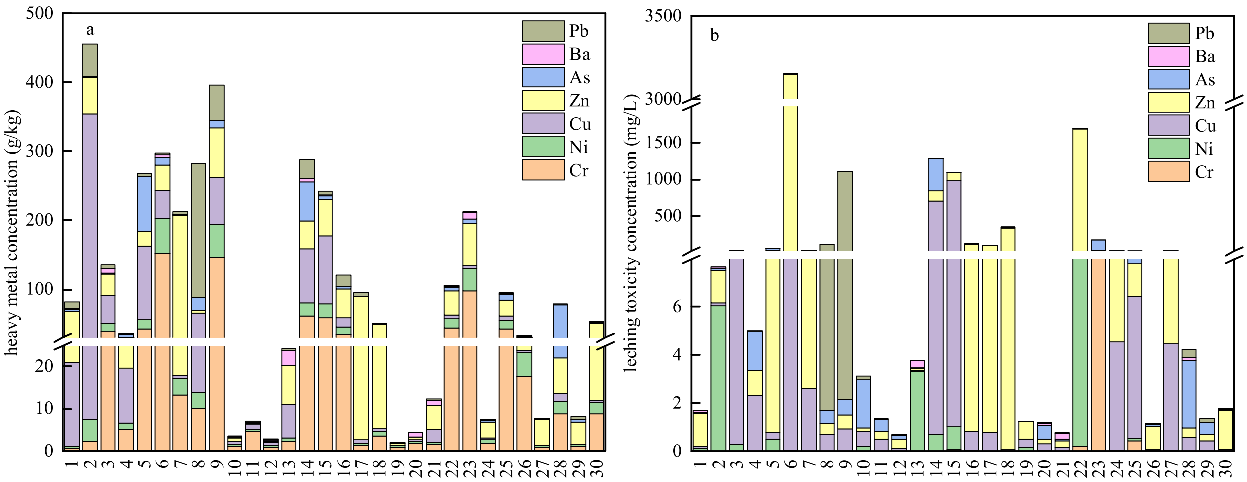

Fig. 5. Heavy metal concentration (a) and leaching toxicity (b) of solid waste from six common generation units.

7778.7 mg/kg. For the wastewater treatment unit, sludges had a high concentration of Zn, which ranged from 5180.07 to 39480.4 mg/kg.

The leaching toxicity concentration of slags from the pyrometallurgy unit and that of sludges from wastewater treatment were relatively low. All samples in these two units were lower than the standard limit in GB 5085.3–2007 [22]. In the roasting and leaching slag from the hydrometallurgy unit, the leaching concentration of Zn was 3141 mg/L, which was higher than the standard limit of 100 mg/L. The leaching concentrations of Pb in copper removal slag and refining slag were 101.75 mg/L and 1108.5 mg/L, respectively, which were higher than the standard limit of 5 mg/L. Five kinds of dusts in the dust collection unit had high leaching concentrations of

Cu, Pb, Zn, and As. In the waste acid treatment unit, the leaching concentration of As in arsenic slag was 141.52 mg/L, which was higher than the standard limit of 5 mg/L.

Because of the high concentration of heavy metals and leaching toxicity, environmental risks should be addressed in the disposal and utilization of solid wastes from the nonferrous smelting industry. Metal recovery technologies for Cu, Pb, Zn, and As should be explored.

3.5. Practical application of the classification method

The new classification method based on a common generation source could be used to guide the disposal and utilization of solid wastes from the nonferrous smelting industry: for slags from pyrometallurgy and hydrometallurgy with a high composition of SiO_2, Fe_2O_3 and CaO could be used as building materials after heavy metal recovery or other methods to reduce heavy metal content [34]. For slags from a waste acid treatment unit, the phase of gypsum also could be used as building material after the removal of As [35]. For dusts from a dust collection unit with a high leaching concentration of Cu, Pb, Zn, and As, if effective metal recovery methods are lacking, landfill disposal would be better after solidification and stabilization [36].

In addition, the classification method offers advantages in identifying the sources of solid wastes from nonferrous smelting. Solid waste with strong alkalinity would come mostly from an electrorefining unit. For solid waste whose mineral phase was mainly calcite would include sludges from a waste water treatment unit.

4. Conclusions

In this study, we categorized solid wastes from the nonferrous smelting industry into six generating units: pyrometallurgy, hydrometallurgy, electrorefining, dust collection, waste acid treatment, and wastewater treatment. We identified similarities among solid wastes from a common generation source and significant differences among solid wastes from different generation sources: slags and sludges from waste acid treatment and wastewater treatment units had a water content of 27.43–52.71% and 51.14–68.27%, respectively, which was significantly higher than those from other metallurgy and dust collection units. The pH of slags from an electrorefining unit was strongly alkaline. The mineral phase of sludges from wastewater treatment was only calcite; slags from a waste acid treatment unit were mainly in the phase of gypsum, claudetite, and anglesite; and the chemical composition of slags from pyrometallurgy and hydrometallurgy units was mainly SiO_2 and Fe_2O_3. Because of the high concentration of heavy metals and leaching toxicity, environmental risks should be addressed in the disposal and utilization of solid wastes from the nonferrous smelting industry. Metal recovery technologies for Cu, Pb, Zn, and As should be explored. This new classification method based on a common generation source would be beneficial to the disposal, utilization, and management of solid waste.

Data availability statement

Data will be made available on request.

CRediT authorship contribution statement

Li Xuebing: Writing – original draft, Methodology, Formal analysis, Data curation. **Yang Yufei:** Supervision, Resources, Conceptualization. **Die Qingqi:** Investigation. **Yang Jinzhong:** Software. **Fanhao Song:** Visualization. **Huang Qifei:** Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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