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Study on leaching characteristics of MSWIFA in different pH environment

Hui Fang, Xin Wu*, Cai Liang and Yongshi Feng

School of Energy and Environment, Southeast University, Nanjing, China

*Corresponding author e-mail: wuxin@seu.edu.cn

Abstract. The change of leaching solution of waste incineration fly ash (MSWIFA) after shaking under different initial extractant pH was studied, and the change trend was compared with that of fly ash under different extractant concentrations. In addition, the leaching characteristics of heavy metals were analyzed under the two methods. The results showed that the pH change of the initial extractant can not truly simulate the environment of fly ash landfill, and the former can not reflect the pH of the final leaching solution. Changing the initial extractant concentration within a certain range can gradually change the final leaching solution from alkaline to acidic, which can truly reflect the environmental changes experienced by fly ash in the landfill (such as rainstorm scouring, drought and other environment). The leaching concentration of heavy metals in the leaching solution obtained by shaking fly ash under different acetic acid concentrations reflects the actual leaching characteristics of heavy metals under different pH conditions, in which Pb and Zn are amphoteric leaching, and the other metals are cationic leaching.

Keywords. MSWI fly ash; initial extractant concentration; initial extractant pH; heavy metal

1. Introduction

At present, the amount of urban waste in China is increasing year by year, which is about 200 million tons by 2021. As one of the important methods of waste treatment and disposal, high-temperature incineration has accounted for one-third of the total waste treatment [1]. The ash generated in the process of waste incineration mainly includes slag and fly ash, of which the amount of fly ash accounts for 2% - 4% of the quality of incineration waste [2]. It is listed in the national list of hazardous wastes because it contains a large amount of toxic and harmful substances such as heavy metals and dioxins [3]. Therefore, incineration fly ash must be solidified / stabilized and meet certain leaching standards before entering the domestic waste landfill plant for regional landfill [4].

As a product of high-temperature incineration, waste fly ash has a complex mineralogical composition. In landfill and other disposal scenarios, the reaction between fly ash and acid and the release of various components are a long-term and slow process. However, the dissolution rate of fly ash is greatly affected by pH value; Therefore, it is necessary to study the degree of reaction between fly ash and acid and the relationship between the release of various elements and pH value under different pH values.

Jiao [5] conducted pH dependent leaching experiments in a wide pH range of 2 to 14, Souhail R [6] used the pH controller to control the initial pH of the extractant to 3, 5, 7, 9 and 11. Cheryl ehalim [7] studied the effects of pH, particle size, leaching duration and liquid-solid ratio on the final leaching concentration of heavy metals. The results showed that environmental factors (such as pH, liquid-solid ratio and ORP) significantly affect the dissolution of Pb and other heavy metals in fly ash minerals [8-10], and pH affects the morphological distribution and solubility of heavy metals in fly ash [11].

However, it is found that the control of pH in different environments in the above experiment only shows a series of changes in pH at the initial time. However, according to the actual situation of fly ash landfill, the environment in which fly ash is located (such as rainstorm and drought) will experience a series of changes from alkaline to acidic [12, 13]. The final pH of leaching solution (leachate in landfill) is the environmental condition, which is the key factor affecting the leaching of heavy metals. Therefore, this paper selected fly ash from waste incineration plant, focused on

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adopting different concentrations of acetic acid to simulate different environmental pH and compared it with the study of changing the pH of initial extractant.

2. Experimental materials and methods

2.1 Experimental materials

The waste fly ash used in the experiment was from waste incineration power plant. X-ray fluorescence spectrum (XRF) analysis was carried out on waste fly ash. The chemical composition of the sample is shown in Table 1. According to table 1, the main component of the content in waste fly ash is CaO, accounting for 42.15% of the total mass of fly ash. This is mainly because the waste incineration power plant uses dry and semi dry methods for tail gas treatment of incineration flue gas, and CaO was injected in this process, resulting in high CaO content in waste fly ash [14].

Table 1. Chemical composition of waste fly ash.

Chemical composition	SiO_2	Al_2O_3	K_2O	Na_2O	CaO	Fe_2O_3	P_2O_5	SO_3	Cl	MgO
Fly ash	4.05	1.24	4.87	12.33	42.15	0.56	0.31	6.67	20.12	1.32

In this experiment, the raw materials were prepared according to the national standard (HJ / T 300-2007) and the liquid-solid ratio was 20. Firstly, a certain amount of waste fly ash were taken in a tray and placed in a constant temperature drying oven to dry at 105 ℃ for 24 hours. 17.25ml glacial acetic acid was taken into a 1L volumetric flask for constant volume.

The research indicated that Pb, Zn, Cu, Cr, CD and other elements in waste fly ash show strong dissolution capacity under acidic conditions [15, 16]. In this experiment, in addition to controlling different initial pH, different concentrations of acetic acid buffer were proportioned, and after 18 h oscillation, heavy metals in the leaching solution were measured by ICP-OES. For comparison, table 2 shows the results of heavy metal digestion of fly ash.

Table 2. Content of heavy metals in washed fly ash after digestion.

Heavy metal	Cr	Cd	Pb	Zn	Cu
Fly ash (mg·kg-1)	26.3	71.8	1865.4	2560.5	274.7
Landfill limit of waste (mg/L)	4.5	0.15	0.25	100	40
Landfill limit of hazardous waste (mg/L)	15	0.6	1.2	120	120

2.2 Experimental methods

After crushing the dried fly ash, it shall be screened through 100 mesh sieve, sealed and stored for standby. The extractants with different pH were prepared with glacial acetic acid and NaOH (superior purity). The initial pH was 3, 5, 7, 9 and 11 respectively. 15g fly ash sample was added into 300ml extraction solution with different pH, then put into the extraction bottle. Placing the extraction bottle on the flip oscillator, and continuously flip and oscillate at the temperature of 23 ℃ at the speed of 30 r / min for 18 hours. After that, the extract was filtered and acidified with nitric acid to pH < 2. The concentration of heavy metals in the solution was determined by inductively coupled plasma emission spectrometer (ICP-OES).

In order to facilitate comparison, acetic acid with different concentrations was also configured in this experiment to simulate different environmental pH. The proportion of materials was shown in table 3. No. 7-12 extractant were put in the full-automatic turnover shaker for shaking for 18 h.

Table 3. Extractant ratio of different concentration.

Number	7	8	9	10	11	12
Concentration of acetic acid (mol/L)	0.3	0.6	0.9	1.2	1.5	1.8
Dosage of acetic acid (mL)	2.4	4.8	7.2	9.6	12	14.4
Deionized water volume (mL)	137.6	135.2	132.8	130.4	128	125.6
Initial pH value	2.76	2.44	2.23	2.07	1.98	1.89

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2.3 Analytical method

The pH is measured by phs-3c pH meter, and the concentrations of heavy metals such as Pb, Zn, Cu, Cr and Cd are measured by ICP-OES. The temperature is 25 ℃, the humidity is 66%, and the concentrations of standard samples are 0, 2, 4, 8 and 16 mg / L respectively.

3. Results and discussions

3.1 Change of pH value of leaching solution

The change of pH of leaching solution with initial pH is shown in Fig. 1. It can be seen from the image that the final pH of the leaching solution is above 12. The pH of the leaching solution first decreased and then increased with the initial acetic acid pH, but there was no significant change. Although the pH of extractant and the content of alkaline substances in fly ash (such as CaO and MgO) would affect the pH value of leaching solution [17, 18], when the content of Cao in fly ash is very high, the latter has a greater impact because it has a certain buffer effect on the pH value. Therefore, the previous leaching experiment on fly ash in a certain pH range cannot really reflect the acid-base environment of the leaching solution.

It can be seen that with the increasing initial concentration of acetic acid, the pH of the leaching solution changes from alkaline to acidic, which is also the real chemical change of fly ash in the specific environment of the landfill (rainstorm scouring and drought). Therefore, in the current situation of high alkalinity of fly ash, only by changing the concentration of acetic acid can the pH value of the final leaching solution show a series of changes.

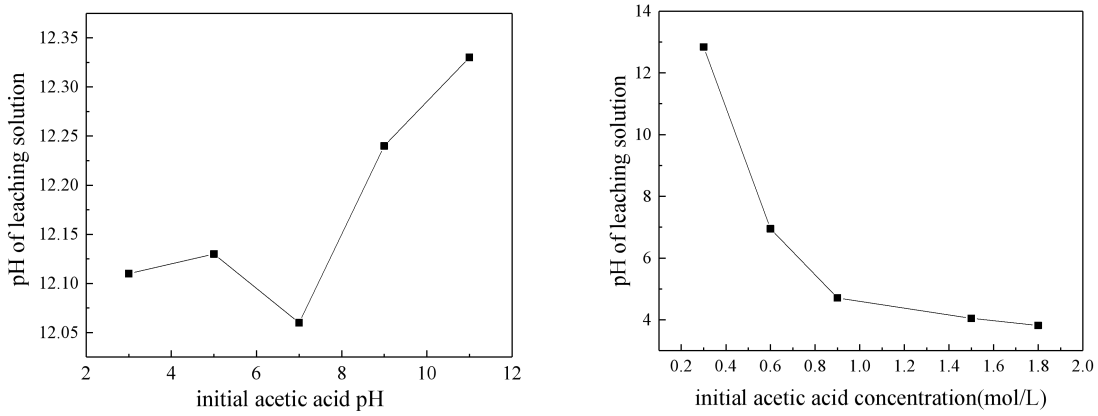

Figure 1. Variation trend of pH of leaching solution with initial acetic acid pH and initial acetic acid concentration.

3.2 Analysis of leaching behavior of heavy metals

The leaching modes of heavy metals in fly ash are generally divided into three types: Cationic type, the leaching concentration decreases with the increase of pH. Anionic type, the leaching concentration increases with the increase of pH. Amphoteric, the leaching concentration is lower under neutral conditions and higher under acidic or alkaline conditions [11].

Studies have shown that Pb and Zn, which were amphoteric metals, can be leached in large quantities under acidic and alkaline conditions [19, 20]. In Fig. 2, the change range of Zn is the largest. Although there is a certain change with the pH of leaching solution, this is because Zn (OH)2 begins to dissolve in alkaline environment to form Zn (OH)42-, Pb is similar to Zn, the release of Pb in the solid phase is mainly caused by the dissolution of Pb (OH)2, which makes the existing form of Pb in the leaching solution almost the same. Under this environment, the leaching

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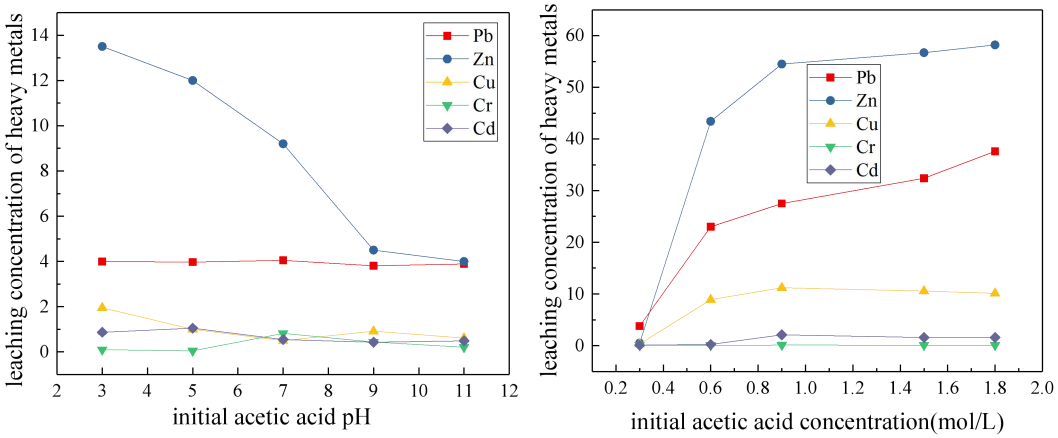
concentration is maintained at about 4 mol/L. In Fig. 2, when the leaching solution changes from alkaline to acidic, the leaching concentrations of Zn and Pb continue to rise, and the peak value is much higher than that in Fig. 2.



(a) (b)

Figure 2. Variation of leaching concentration of heavy metal with pH of leaching solution under different methods.

In Fig. 2, the leaching concentration of Cu, Cr and CD is always low, generally lower than 2 mol/L, because the final pH is alkaline and metal ions form precipitation with (OH)2-. The three metals gradually began to dissolve in acid when the initial acetic acid concentration was high, forming free metal ions, in which Cu2+ was close to 15 mol/L in strong acid environment. Therefore, it can be found that changing the initial acetic acid pH to simulate a series of environmental changes is more scientific and reasonable than previous studies.

4. Conclusion

Previous experiments on a series of changes in the pH of the initial extractant cannot truly simulate the environment of the fly ash landfill. The former cannot reflect the pH of the final leaching solution. After 18 h vibration, it is still alkaline and the pH is finally stable at 12-13.

Changing the initial extractant concentration within a certain range can gradually change the final leaching solution from alkaline to acidic, which can more truly reflect the environmental changes experienced by fly ash in the landfill.

The leaching concentration of heavy metals in the leaching solution obtained by shaking fly ash under different acetic acid concentrations reflects the actual leaching characteristics of different heavy metals under different pH conditions.

Acknowledgments

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