

HEAVY METALS LEACHING BEHAVIOURS IN MSWI FLY ASH STABILIZED BY AN ORGANIC CHELATING AGENT

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INTRODUCTION

Municipal solid waste incineration fly ash (MSWIFA) is a secondary pollutant produced during municipal solid waste incineration and is classified as a hazardous waste because of its rich content of heavy metals (Quina et al., 2011). Considering the increasing production of fly ash leading to excessive pressure on safety landfills, the government stipulates that treated MSWI fly ash can enter sanitary landfill after meeting the requirements of the “Standard for pollution control on the landfill site of municipal solid waste” (GB16889-2008). Treatment technologies for fly ash include separation, solidification/stabilization, thermal treatment and reutilization (Wang et al., 2021). Stabilization is widely used in China by virtue of its simplicity of operation, high stabilization efficiency and no increase in fly ash volume (Li et al., 2019).

In sanitary landfills, fly ash is often landfilled separately from municipal solid waste. This means the fly ash entering the landfill is not exposed to the environment of organic acid leaching, but mainly rainwater leaching (Li et al., 2020). Therefore, in this study, in addition to the original provisions, the leaching behavior of stabilized fly ash in simulated acid rain was also investigated.

MATERIALS AND METHODS

MSWI fly ash used in this experiment was obtained from a waste incineration plant in Chengdu, Sichuan. The chelating agent used was piperazine chelating agent (TS300) from Tosoh, Japan. 100 g fly ash was mixed with 1%, 3%, 5%, 7% and 10% (w/w) of chelating agent, stirred for 10 min under a stirrer, stored in sealed bags, and sampled at 0d, 14d and 28d for leaching experiments. The leaching toxicity of stabilized fly ash entering the landfill under municipal solid waste leachate and acid rain was simulated using the acetic acid buffer solution method (HJ/T 300-2007) and the Sulphuric acid & nitric acid method (HJ/T 299-2007).

A modified four-step procedure sequential BCR extraction method was used to analyze heavy metal speciation in fly ash, namely, acid soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3), and residual fraction (F4). The acid soluble fraction and

reducible fraction are considered as unstable speciation. the oxidizable fraction and residual fraction are considered as stable speciation.

RESULT AND DISCUSSION

Due to the imperfect classification and recycling system of municipal solid waste in China, municipal solid waste is mixed with some waste containing heavy metals, resulting in a large amount of Zn, Pb, Cd and other heavy metals in fly ash. In the acetic acid buffer solution method (HJ/T 300-2007), the leaching amounts of Pb and Cd in raw fly ash are 1.04 mg/L and 5.39 mg/L, which far exceed the limits of 0.25 mg/L and 0.15 mg/L specified in the GB16889-2008. So the raw fly ash needs to be treated and meet the standards before entering the sanitary landfill.

The leaching concentrations of Pb and Cd in acetic acid solution treated with different TS300 dosages are shown in Figure 1. With the increase of TS300 dosage, Pb and Cd are stabilized in fly ash, which leads to the decrease of their leaching in acetic acid solution. When the dosage of TS300 is 3%, the leaching concentrations of Pb and Cd in acetic acid leaching solution are 0.06 mg/L and 0.06 mg/L respectively, which are lower than the limit in GB16889-2008. However, there is still the possibility of re-dissolution of heavy metals in fly ash stabilized by chelating agents. After storage for 14 days, the leaching concentrations of Pb and Cd of fly ash stabilized are 0.10 mg/L and 0.17 mg/L, respectively, and increased to 0.23 mg/L and 0.62 mg/L, respectively, after storage for 28 days. Obviously, Cd exceeds the limit value in GB16889-2008 after 14d storage. Because the stabilized fly ash is oxidized by the air, which causes the decomposition of the stable heavy metal fraction, and the release of Pb and Cd leads to its easy transfer to the environment (Sakanakura, 2007).

The leaching concentration of Pb from raw fly ash in acid rain is 11.71 mg/L, which is much higher than that in acetic acid, and more than 46 times the limit value in GB16889-2008. Therefore, under the simulated acid rain scenario, Pb is a key heavy metal to dealt with. As shown in Figure 2, when the addition of chelating agent is 3%, the leaching concentration of Pb in simulated acid rain

(0.016 mg/L) can reach the limit value. With the extension of storage time, Pb has a tendency to be re-released, but it is not as obvious as in acetic acid leaching.

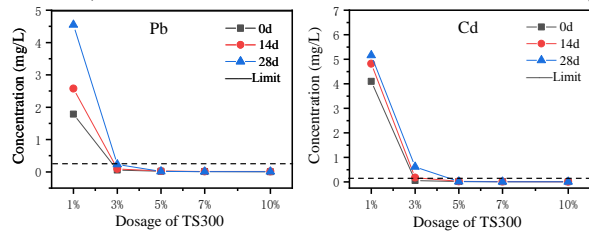


Figure 1. The leaching concentrations of Pb and Cd in acetic acid solution treated with different TS300 dosages

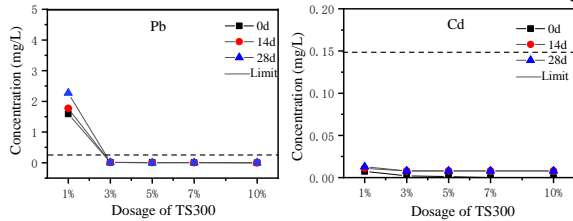


Figure 2. The leaching concentrations of Pb and Cd in acid rain solution treated with different TS300 dosages

As shown in Figure 3, the speciation of heavy metals (e.g. Pb, Cd) is changed after the addition of chelating agents. In raw fly ash, the speciation proportion of Pb follows the order of oxidizable fraction (42.66%) > reducible fraction (42.14%) > residual fraction (13.78%) > acid soluble fraction (1.41%). This is the reason why the large leaching of Pb from fly ash in acetic acid leachate and sulfuric acid nitric acid leachate. After the addition of chelating agent, the acid soluble fraction decreases to less than 1% and the reducible fraction also decreases. When the addition of chelating agent increases to 3%, the sum of acid soluble fraction and reducible fraction decreases to 0.64%, and most of Pb belong to the stable fraction and are not easy to leach. However, with the extension of storage time, the sum of acid soluble fraction and reducible fraction increases to 1.09%, and the leachable fraction increased. This is the same phenomenon as it showed in Figures 1 and 2.

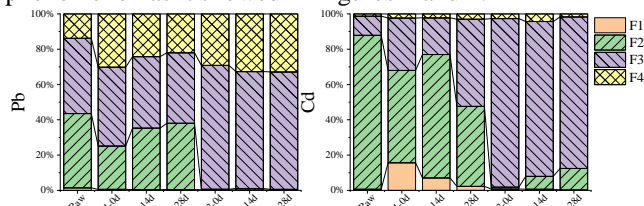


Figure 3. Proportion of Pb and Cd speciation in the FA sample determined by BCR

In raw fly ash, Cd mainly exists in the unstable reducible fraction (87.22%), while the oxidized fraction (10.86%) and residual fraction (1.24%) are very small. The addition of chelating agent has obvious stabilization effect on Cd. When the chelating agent dosage increases to 3%, the reducible state of Cd decreases from 87.22% to 1.07%. With the extension of storage time, the sum of acid soluble fraction and reducible fraction increases to

7.93% (14d) and 12.42% (28d). Therefore, chelating agents stabilize the heavy metals by combining the chelating groups with the heavy metals to reduce their mobility (Wang et al., 2015).

CONCLUSION

This study shows that when the chelating agent dosage is 3%, a large number of Pb and Cd in fly ash are stabilized, but with the increase of storage time, the stabilized Pb and Cd are re-released. According to the analysis of BCR method, chelating agent can change the speciation of heavy metal, and transformed it from unstable fraction (F1 and F2) to stable fraction (F3 and F4). Because chelating groups can effectively bind to heavy metals, reducing their mobility. Therefore, chelating agents have great potential in treating heavy metals in fly ash and reducing their toxicity.

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