

THE LONG-TERM PERFORMANCE OF CONCRETE AMENDED WITH MUNICIPAL SEWAGE SLUDGE INCINERATION ASH

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ABSTRACT

Municipal sewage sludge incineration ash (MSSIA) is commonly used as a raw material in concrete, but few studies have focused on the long-term environmental tolerance of municipal sewage sludge incineration ash concrete (MSSIAC). In this study, acid rain leaching, salt erosion, and freezing-thawing tests were used to evaluate the long-term environmental behavior of the optimal MSSIAC formulation. One optimal MSSIAC formulation replaces 5% MSSIA, and results in a 7.2% increase in unconfined compressive strength (UCS), indicating that MSSIA-amended concrete can have enhanced UCS. Only Zn was released from the optimal MSSIAC formulations (replacing 5% or 10% MSSIA) in the acid rain leaching test, but only up to 0.071 mg Zn L⁻¹, which was within the acceptable environmental range. No heavy metals were released at concentrations above the method detection limits in the salt erosion and freezing-thawing tests. The optimal MSSIAC formulations have excellent long-term environmental tolerance while meeting the requirement of UCS, and provide a valuable use for MSSIA.

Keywords: Concrete; Municipal sewage sludge incineration ash; Long-term; Environmental tolerance; Heavy metal

INTRODUCTION

With rapid economic development and increase in the standard of living, municipal sewage sludge generation is continuously escalating. According to the statistics, dry municipal sewage sludge produced in the municipal sewage treatment process in China exceeded 12 million metric tonnes in 2017 (Z. Chang et al., 2020; S. Ledakowicz et al., 2019), and only about 20% was disposed of in harmless ways such as the production of construction materials, thermal treatment (P. Das et al., 2020), agricultural application (M. Skowrońska et al., 2020), and sanitary landfill (A. Kelessidis and A.S. Stasinakis, 2012). Among them, incineration technology provides the advantages of volume reduction, sterilization, and electricity generation (O. Krüger and C. Adam, 2015); however, the municipal sewage sludge incineration ash (MSSIA) generated

after incineration requires landfill disposal and can possibly cause secondary pollution such as the release of heavy metals. Therefore, MSSIA treatment merits global concern and research.

Unlike fly ash from the hazardous waste incineration process, MSSIA is a general industrial solid waste, although it contains certain heavy metals. Recently, MSSIA disposal methods have been reported, including landfills (S. Donatello et al., 2010), stabilization and solidification (R. Siddique, 2010), which can reduce the potential risks of MSSIA. However, MSSIA disposal will still require extensive land resources after treatment. MSSIA can also cause secondary pollution after long-term environmental exposure because of the limited immobilization of heavy metals. It still needs more thoughtful strategy for MSSIA disposal.

Concrete is in high demand for infrastructure construction. Because MSSIA has pozzolanic activity (L. Chen and D.-F. Lin, 2009), it can be used as raw material for concrete, which consumes the MSSIA and provides a valuable use for this waste product. This is a method that combines both waste management and resource utilization. Compared with other treatment methods, concrete prepared by ash incineration has obvious advantages in resource utilization. Therefore, this method has received extensive research and attention.

MSSIA has been used as cementitious material to replace about 10–20 % of cement in concrete, while having no significant impact on the concrete strength (G. Rutkowska et al., 2018; Z. Chen and C.S. Poon, 2017; Z. Chen et al., 2018). Municipal sewage sludge incineration ash concrete (MSSIAC) can meet strength requirements, and most published studies focus only on physical properties such as unconfined compressive strength (UCS) and flexural strength to characterize the effect of MSSIA on concrete (S. Chakraborty, 2017). Though some environmental tolerance studies on MSSIAC involve leaching test, these only reflect the short-term environmental effects, not long-term effects (J.-s. Li et al., 2017; C.J. Lynn et al., 2018). In some areas, high rainfall occurs, with a high frequency of acid rain, increasing the requirements for environmental

tolerance. Short-term leaching test cannot fully reflect the environmental performance of MSSIA, and the long-term environmental behavior must be studied. In addition, the changing global climate may also increase the risk of release of pollutants from MSSIA, and it is important to explore the long-term performance of MSSIA under extreme environmental conditions.

In this study, MSSIA was introduced into concrete to find an optimal concrete formulation for MSSIA disposal. The MSSIA was then evaluated in severe long-term environment exposure scenarios. It aims to provide a feasible and safe disposal strategy for MSSIA.

MATERIALS AND METHODS

Materials and sample preparation

MSSIA (100 kg) was obtained from a municipal sewage sludge incineration plant located in Hangzhou, China. This incineration plant uses circulating fluidized bed sludge incineration at a capacity of 500 t d⁻¹.

Cement used in this study was P.O. 42.5. The sand and stone were purchased from a quarry in Hangzhou, China. The stone was sifted into five grain-grades: 16–19 mm, 13.2–16 mm, 9.5–13.2 mm, 4.75–9.5 mm, and 2.36–4.75 mm. Sand was sifted to 0.075–2.36 mm. Concrete was prepared according to standard (EN 12390-2, 2000) by mixing cement, stone, sand, MSSIA (except in the controls), and water and curing the mixture for up to 28 d, either in a 100 mm × 100 mm × 100 mm cubic concrete form or a 60 mm × 60 mm × 150 mm cylindrical concrete form.

A simulated acid rain solution was prepared based on the acid rain chemistry in China, containing 3.10 mg L⁻¹ CaCl₂, 2.80 g L⁻¹ NH₄Cl, 0.91 g L⁻¹ NaCl, and 0.75 g L⁻¹ KCl with a pH of 4.00 and a molar ratio of SO₄²⁻: NO₃⁻ of 4.88: 1. These proportion and ratio are some of the harsh conditions (not the harshest) seen many times in the acid rain in southern Chinese cities. We chose these conditions to verify the environmental tolerance of MSSIA in harsh environments. A simulated seawater solution was prepared based on the actual salinity of offshore seawater, containing 3.5 % NaCl (w/w).

Experimental

The MSSIA was set with reference to standard (EN 12390-2, 2000). The tested groups using MSSIA to replace sand were designed as shown in Table 1. Namely, seven groups of MSSIA with different proportions of MSSIA were set. Each group of MSSIA was cured in a 100 mm × 100 mm × 100 mm cubic concrete form. It is worth mentioning that, simply using MSSIA to replace sand without adding water will affect the fluidity of cement. Therefore, we added a certain proportion of water to keep the fluidity consistent. Four replicates were used for each group. All groups of MSSIA were aged at room temperature.

After 28 d, the UCS was calculated with reference to standard (EN 12390-3, 2001).

Table 1 Content of each component in MSSIA tested groups

| Groups | Water | Cement | Sand | Stone | MSSIA |
|--------|-------|--------|------|-------|-------|
| C-0 | 9.0 | 22.4 | 22.0 | 46.7 | 0 |
| C-5 | 9.2 | 22.4 | 20.9 | 46.7 | 0.84 |
| C-10 | 9.5 | 22.4 | 19.8 | 46.7 | 1.68 |
| C-15 | 9.7 | 22.4 | 18.7 | 46.7 | 2.52 |
| C-20 | 10.0 | 22.4 | 17.6 | 46.7 | 3.36 |
| C-25 | 10.2 | 22.4 | 16.5 | 46.7 | 4.2 |
| C-50 | 11.5 | 22.4 | 11.0 | 46.7 | 8.4 |

Unit: w/w, %.

Based on the UCS results, the optimal MSSIA formulation was selected for further tests of environmental tolerance under scenarios of simulated acid rain leaching, salt erosion, and freezing-thawing. According to the actual height of concrete pavement in China, 180 mm was selected as the height of the specimen for the simulated acid rain leaching test and salt erosion test.

The simulated acid rain leaching test were carried out in leaching columns (61 mm diameter, 180 mm height) with a spraying set-up that included a 25 L water tank and a drop filter. The optimal cylindrical MSSIA was placed into the leaching columns, and simulated acid rain was continuously sprayed over the MSSIA by controlling the drop filter. To mimic the most severe scenarios, the leaching experiment lasted for 60 d, which equaled 100 y of rainfall.

The salt erosion test was carried out in rectangular containers (80 mm length and width, 180 mm height). The optimal cylindrical MSSIA was placed into the containers filled with simulated seawater, and sealed. The erosion experiment lasted for 42 d.

The freezing-thawing test was carried out in cylindrical containers (200 mm diameter, 110 mm height). The optimal cubic MSSIA was placed into the containers filled with water. Each freezing-thawing cycle consisted of freezing at -20 °C for 12 h and thawing for 12 h at 25 °C, and the freezing-thawing experiment lasted for 42 cycles.

Analyses

The MSSIA moisture content and organic fraction was measured by standard methods (L. Hu et al., 2019). Heavy metals, including Cu, Pb, Cr, Cd, Ni, and Zn, were measured by atomic absorption spectroscopy (AAS, ZEE nit 700p, Germany) after acid (HCl-HNO₃-HF-HClO₄) digestion (J. Yao et al., 2017).

The MSSIA mineral and elemental compositions were determined by X-ray diffraction (XRD, Rigaku D/Max-III A, Japan) and X-ray fluorescence (XRF, Axios-Advanced, Netherlands). The UCS of MSSIA was measured according to standard EN 12390-3 using a hydraulic test machine. Each test was performed in triplicate, excluding the XRD and XRF.

Statistical analysis was performed with SPSS 22.0 software, and the standard deviation was obtained by descriptive statistics. A correlation analysis was performed using Tukey's test. Differences were considered to be statistically significant when $p < 0.05$.

RESULTS AND DISCUSSION

Characteristics of MSSIA and MSSIA/C

The mineral and elemental compositions of MSSIA obtained from the XRD was shown in Fig. 1. The major mineral components in MSSIA were quartz and anhydrite, similar to coal ash and cement. The major components of MSSIA were SiO_2 , Al_2O_3 , Fe_2O_3 , and CaO , with proportions of 10.3 %, 5.04 %, 15.4 %, and 34.1 %, respectively; these compounds are the main reactive components of pozzolan. It has been previously shown that MSSIA has pozzolanic activity, and the addition of MSSIA to concrete may increase concrete strength (Q. Wang et al., 2012).

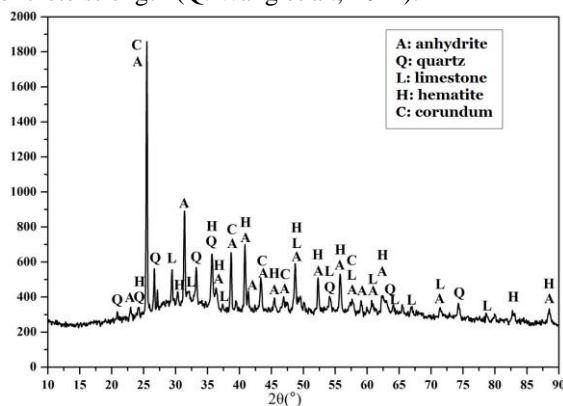


Fig. 1. XRD result of MSSIA.

In addition, decreasing the chloride and bromine contents in concrete can prevent damage caused by reinforcement corrosion (A.A. Naqvi et al., 2015; A.A. Naqvi et al., 2006). The contents of Cl and Br in MSSIA are 0.271 % and 0.026 % respectively, which indicates the MSSIA does not require pretreatment to reduce the corrosion risk. The results demonstrate that MSSIA not only has the mineral and elemental components similar to coal ash and cement and pozzolanic activity, but it also has low Cl and Br content and poses almost no corrosion risk to steel bars. Therefore, MSSIA has the potential to replace cement or aggregate raw materials and may be an ideal raw material for concrete.

With increasing amounts of MSSIA, the UCS increased

then decreased (Fig. 2). With a MSSIA replacement rate of 5 %, sample C-5 has the highest UCS of 41.8 MPa, which is 7.2 % higher than the control sample, indicating that a small amount of MSSIA can increase UCS of concrete because of its pozzolanic activity (X. Xie et al., 2019; N. Saboo et al., 2019). With a MSSIA replacement rate of 10%, the UCS of C-10 was 32.7 MPa, 16.2 % lower than control sample, possibly because the increased proportion of MSSIA diluted the cement and hindered its normal cementation (M. Khan and M. Ali, 2019). However, the UCS of C-10 could still meet the requirements of commercial application. Higher proportions of MSSIA resulted in a further decrease in UCS, and at replacement rate greater than 15% (C-15 and above), the strength requirements for commercial applications were no longer met.

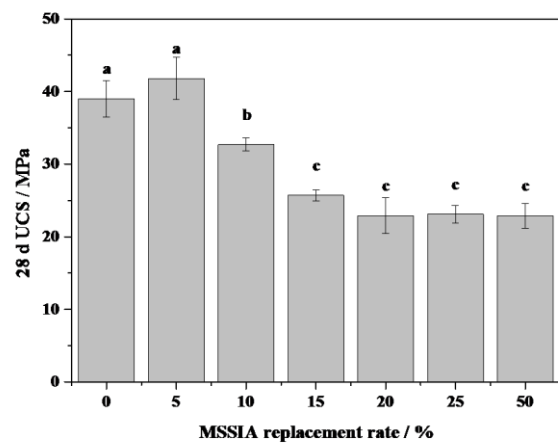


Fig. 2. UCS results of MSSIA/C tested groups (The different letter indicates significant different at 0.05 level).

The sample C-5 has the highest UCS. Although the UCS of sample C-10 is hard compete with C-5, but it has a larger MSSIA capacity, which is good for the environment, and it maintains a commercially viable strength. Therefore, the samples C-5 and C-10 represented the optimal MSSIA/C formulations. These samples balanced the required UCS and incorporation of some proportion of MSSIA, providing high application value.

Long - term environmental tolerance of optimal MSSIA/C

To evaluate the long-term environmental tolerance of the optimal MSSIA/C samples (C-5 and C-10), simulated acid rain leaching, salt erosion, and freezing-thawing were tested. C-0, containing no MSSIA, represented the control group. The Cu, Pb, Cr, Cd, Ni, and Zn contents of MSSIA were 4652 mg kg^{-1} , 600 mg kg^{-1} , 11.1 mg kg^{-1} , 0.00, 177 mg kg^{-1} , and 406 mg kg^{-1} , respectively. And the content of heavy metals in C-5 and C-10 are shown in Table 2.

Table 2 Content of heavy metals in C-5 and C-10

| Heavy metals | Zn | Cr | Cd | Pb | Ni | Cu |
|--------------|------|------|-------|------|------|------|
| C-5 | 39.1 | 5.04 | 0.093 | 0.00 | 1.49 | 3.41 |
| C-10 | 78.2 | 10.1 | 0.186 | 0.00 | 2.97 | 6.82 |

Unit: mg kg⁻¹.

The content of heavy metals in the acid rain leaching tests of the control, C-5, and C-10 samples are shown in Fig. 3a, 3b, and 3c, respectively. No heavy metals except Zn were detected in the leachate representing 100 y of acid rain, which could be because MSSIA has a higher concentration of Zn compared with other heavy metals, but could also be a result of the solidification mechanism of cement (W. Ma et al., 2019). The cementitious components in cement can combine with heavy metals during solidification, and Zn is more reactive and more easily released under acidic conditions compared with other heavy metals. The results from the acid rain leaching test also show that MSSIA can stabilize heavy metals, and the environmental risk of release of heavy metals from MSSIA through acid rain leaching is low.

The concentration of released Zn reached maximum values of 0.066 mg L⁻¹ on year 55 of the acid rain leaching test for C-0, 0.071 mg L⁻¹ on year 5 for C-5, and 0.070 mg L⁻¹ on year 5 for C-10. The experimental groups (C-5 and C-10) released the highest Zn concentrations at the first sampling point, which may be because some metal ions that were not completely solidified were released at the first sampling point. After this, the release of Zn slowed and the concentration fluctuated in a lower range. There was no significant difference in the concentration of Zn released by the three MSSIA groups, and the concentration of Zn released was very low under all conditions and almost equal to that of normal surface water. These results demonstrate that acid rain will not threaten the stability of MSSIA.

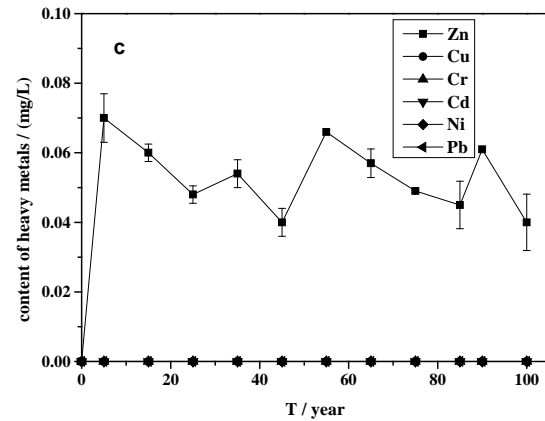
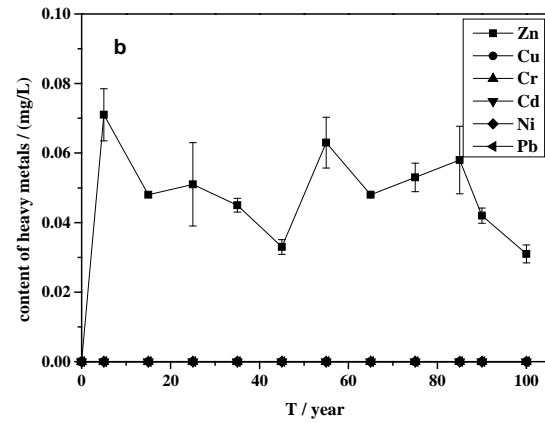
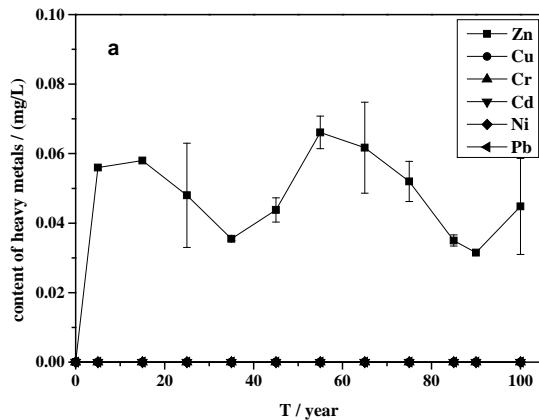


Fig. 3. The heavy metals contents in acid rain leaching test result of MSSIA (a, b, and c mean the control, the C-5, and the C-10, respectively)

The accumulated Zn amounts for the three test groups is shown in Fig. 4a. There was no significant difference between the accumulated Zn amounts for the control and C-5 group, because of the low initial concentration of heavy metals and the stable solidification effect. C-10 group containing a higher amount of MSSIA so that C-10 had a higher accumulated Zn amount compared with the control. The source of the additional Zn is the MSSIA addition. Therefore, the ratio of the extra Zn released to the total C-10 content were calculated, and a linear curve was used to fit the cumulative ratio in Fig. 4b, which shows that it may take approximately 9,076 years for the Zn to be fully released. The service life of ordinary Portland concrete is about 120 years (S. Demis and V.G. Papadakis, 2019), and MSSIA will have a low heavy metal release and excellent environmental safety during this period. Even after the approximate 9,076 years, after the Zn is fully released, the leachate concentration remains at a safe level, below the highest level of EU surface water quality standards. Therefore, MSSIA has excellent long-term environmental tolerance under simulated acid rain leaching conditions.

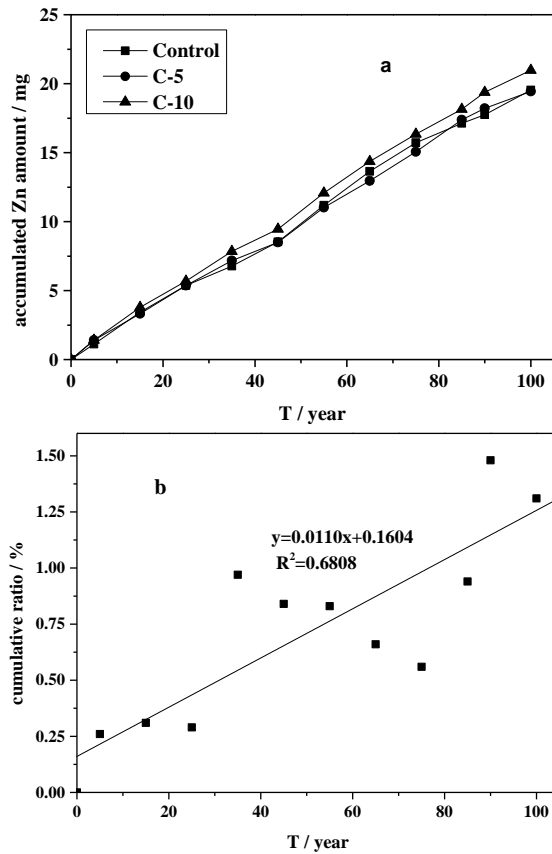


Fig. 4. a, the accumulated Zn amount from three MSSIA groups; b, the cumulative ratio of the extra Zn released to total C-10 content.

The impact of salt erosion and freezing-thawing scenarios on MSSIA results are shown in Table 3. The high salt content of seawater can erode the concrete structure, and the concrete may start to release heavy metals (V. Marcos-Meson et al., 2018). The freezing-thawing process freezes the free water in the pores of concrete, causing volume expansion and can damage the internal structure of concrete, also causing the concrete to release heavy metals (M.K. Ismail and A.A.A. Hassan, 2019). In this study, all heavy metal concentrations were below the method detection limit in both the salt erosion and freezing-thawing tests (Table 3). The heavy metals may have been immobilized in the concrete in very stable states, such as residual states. Salt erosion and freezing-thawing, which damage the physical structure of the concrete, may only release the heavy metals in less stable states, without affecting the heavy metals immobilized in stable states. These results indicate that MSSIA can effectively stabilize heavy metals, and protect them from release under extreme environmental conditions such as salt erosion and freezing-thawing, adding MSSIA does not increase the risk of heavy metal release from concrete.

Table 3 Salt erosion and freezing-thawing test results

| Salt erosion test | | | | | | |
|-----------------------|-----------------|----|----|----|----|----|
| Days | Zn | Cr | Cd | Pb | Ni | Cu |
| 0 | ND ^a | ND | ND | ND | ND | ND |
| 7 | ND | ND | ND | ND | ND | ND |
| 14 | ND | ND | ND | ND | ND | ND |
| 21 | ND | ND | ND | ND | ND | ND |
| 28 | ND | ND | ND | ND | ND | ND |
| 35 | ND | ND | ND | ND | ND | ND |
| 42 | ND | ND | ND | ND | ND | ND |
| Freezing-thawing test | | | | | | |
| Days | Zn | Cr | Cd | Pb | Ni | Cu |
| 0 | ND | ND | ND | ND | ND | ND |
| 7 | ND | ND | ND | ND | ND | ND |
| 14 | ND | ND | ND | ND | ND | ND |
| 21 | ND | ND | ND | ND | ND | ND |
| 28 | ND | ND | ND | ND | ND | ND |
| 35 | ND | ND | ND | ND | ND | ND |
| 42 | ND | ND | ND | ND | ND | ND |

a: ND means not detection.

CONCLUSIONS

As the proportion of MSSIA in concrete increased, the MSSIA UCS first increased and then decreased. A replacement of 5% MSSIA resulted in a 7.2% increase in UCS (C-5), while a replacement of 10% MSSIA resulted in a 16.2% decrease in UCS (C-10). Only Zn was released from the optimal MSSIA formulations (C-5 and C-10) in the acid rain leaching test, but at its highest concentration, only 0.071 mg L^{-1} was detected, within the acceptable range. No heavy metal release concentrations were above the method detection limit in both the salt erosion and freezing-thawing tests. The optimal MSSIA has excellent long-term environmental tolerance while meeting the requirement of UCS, and represents a valuable use of MSSIA.

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