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Cement-Based Solidification of Incinerated Sewage Sludge Ash by the Addition of a Novel Solidifying Aid

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Abstract

The effects of a novel solidifying aid on the solidification of incinerated sludge ash were investigated in this study. The compressive strength and the heavy metal leaching toxicity of the solidification block were measured, and the composition and the microstructure were also detected by XRD and SEM. The results showed that the optimal solidifying agent was as follows: incinerated sewage sludge ash (ISSA): Portland cement: Kaolin: solidifying aid= 100:40:10: 0.7. The compressive strength of 12.74 MPa was observed when ISSA was mixed with the best solidification condition after 28 days curing. TCLP test results showed that the concentrations of all the metals in the leachate of the solidified samples are below those set in the maximum solubility limit issued by the Environmental Protection Agency in China. The XRD and SEM analysis indicated that the structure of the solidification block was of many acicular crystals and very dense. Furthermore, quartz, $CaAl_2Si_2O_8$, $Ca_2Al_2SiO_7$ and other materials could be found in the solidification blocks, which were known as improving the compressive strength of the solidification blocks.

Keywords: Incinerated sewage sludge ash (ISSA); Solidification; Compressive strength; Solidifying aid

Introduction

The limited space and the high cost of land disposal led to the development of recycling technologies and the reuse of sewage sludge ash in structural and construction materials [1]. Incineration, as a method of solid waste management, could reduce the sludge volume by 90-95%, has become an important method for disposal sewage sludge following the EU wide ban on sea disposal in 1998 [2]. It could generate solid residues, such as bottom and fly ash as well as off-gas cleaning residues with high levels of heavy metals, inorganic salts and other organic compounds. It is estimated that 1.2 million tonnes of incinerated sewage sludge ash (ISSA) are currently produced annually in the EU and North America [3], and a further 0.5 million tonnes/yr in Japan alone [4]. For this reason it requires special management. In recent years, ISSA has been widely used in the production of construction material [5], concrete and cement [6,7], tile [8,9], pavement materials [10], and lightweight aggregate [11,12]. The alternative utilization of ISSA used as construction material according to its cementitious properties by the method of solidification was reported little. It basically involves waste containment within a solid matrix using different binder materials such as cement, pozzolans, clay and Kaolin [13]. It is a relatively new treatment process that has the potential to reduce leachability of hazardous constituents from the disposed waste and the research work on solidification of ISSA has not been extensive. In this study, an unburned solidification technology was applied to the treatment of sewage sludge ash with the use of different types of solidifying agent and aid. The objective of this study was to optimize the solidification agent based on the Portland cement and a new solidifying aid, and analyze solidification mechanism by using X-ray diffraction (XRD) and scanning electron microscope (SEM) techniques. Furthermore, leaching tests was examined to investigate the risk of hazardous heavy metals leaching following the use of solidified sewage sludge ash as construction materials.

Materials and Methods

Materials

The incinerated sewage sludge ash (ISSA) used in this research work was collected from TaoPu Wastewater Treatment Plant located in PuTuo District, Shanghai, China. Commercial Portland cement (PC) typed CEM I 42.5 was used in the experiments as the main binder. The Kaolin used in this research was typed SD-C6 with 300 meshes. Both of their chemical and physical properties were shown in Tables 1 and 2. The solidifying aid (SS-SA) used in this study was a mixture of H_2SO_4, H_3PO_4, and Na_2SiO_3, which mass ratio was H_2SO_4:H_3PO_4:Na_2SiO_3=1:8:91.

Sample preparation

Twenty-three treatments were shown in Table 3, the dosages of the components were determined according to the previous experiments. All the samples were prepared by mixing the incinerated sewage sludge ash and solidification agents together with distilled water. The water cement ratio was fixed at 0.35. The mortar mixtures were mixed following the standard test method ASTM C 305-94 to achieve a uniform distribution of the mixtures before transferred to the 40 mm×40 mm×40 mm mould [14]. The mortar mixtures were cast in moulds for the first 24 h at room temperature and then the blocks were demolded and set in a temperature- controlled curing box at 25±1°C for 7, 14 and 28 days to determine the optimum solidification agent formula.

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'%wt	SiO_2	Fe_2O_3	Al_2O_3	TiO_2	CaO	MgO	SO_3	P_2O_5	K_2O	Na_2O	Total
ISSA	40.87	9.63	16.89	0.69	11.66	2.25	5.03	8.19	1.69	1.01	98.85
Cement	20.30	3.56	7.65	0.32	61.25	2.13	1.98	-	1.02	0.36	100.97
Kaolin	54.96	0.47	42.35	0.09	0.52	0.42	0.19	-	0.53	0.32	99.86

Table 1: Chemical compositions of materials used in experiment.

	Fineness (m^2/kg)	Density (kg/m^3)
ISSA	1980	1.56
Cement	347	3.15
Kaolin	524	2.43

Table 2: Physical properties of materials used in experiment.

		Solidified agent			
Group	No.	ISSA	Cement	kaolin	Solidifying Aid
G1 ISSA/PC	1	100	0	10	0
	2	100	10	10	0
	3	100	20	10	0
	4	100	30	10	0
	5	100	40	10	0
	6	100	50	10	0
	7	100	60	10	0
G2 ISSA/PC/ Kaoline	8	100	40	0	0
	9	100	40	10	0
	10	100	40	20	0
	11	100	40	30	0
	12	100	40	40	0
	13	100	40	50	0
	14	100	40	60	0
	15	100	40	70	0
G3 ISSA/PC/ Kaoline /SS-SA	16	100	40	10	0
	17	100	40	10	0.1
	18	100	40	10	0.3
	19	100	40	10	0.5
	20	100	40	10	0.7
	21	100	40	10	1.0
	22	100	40	10	1.5
	23	100	40	10	5.0

Table 3: Mixture proportions (wt%).

Compressive strength test

The physical strength of the solidified matrix is significant since it determines the suitability of the solids to be used as construction material and for secure landfill stacking. Samples curing for 7 days, 14 days and 28 days were tested by determination of compressive strength according to the methods described by the European Standard EN 196.1 [15]. For each mortar and curing age, three specimens were tested.

Toxicity characteristic leaching procedure (TCLP)

The leaching test for heavy metals from the solidified blocks was assessed using the toxicity characteristic leaching procedure (TCLP) as defined by the U.S. EPA on the samples cured under 25 ± 1°C for 28 days [16]. The TCLP test was carried out by crushing the sample into powder using a mortar and pestle to reduce the particle size to smaller than 9.0 μm with 0.1M acetic acid and 0.0643 M NaOH solutions (pH 2.8), at a liquid/solid ratio of 20:1. The extraction vessels were rotated at 120 rpm for 20 h (TDL-5-A, Anke Ltd,Shanghai). The leachate was filtered through a 0.45 μm membrane filter to remove suspended solids and then divided into two portions, one of which was used for pH measurement and the other for the determination of metals present in the leachate by ICP-AES (Iris Advantage, Thermo Electron Corporation,US) according to the standard methods. Each extraction was done in triplicate, and the average value was reported to ensure the reproducibility of the data.

XRD and SEM analysis

In order to investigate the influences of different mix ratios on specimen properties, chemical compositions were analyzed using X-ray diffraction (D/max-2200/PC, Rigaku Corporation,Japan) and sample microstructures were studied using LV UHR FE-Scanning Electron Microscopy (Na- no SEM 230, NOVA FEI, US) to microscopically analyze the physicochemical variations of the ISSA before and after the solidification in terms of the strength-enhancing effects of the solidifying reaction. The XRD and SEM analyses were conducted for each of the solidified samples, which were prepared by mixing fixed proportions of ISSA/solidifying agent/solidifying aid and then curing them under 20± 1°C for 28 days. An X-ray diffraction analyzer was used to investigate the constituents of solidified sludge ash samples at the 30 kV of acceleration voltage condition. SEM imaging was performed to obtain the micrographs of the sludge samples before and after solidification at 20 kV of acceleration voltage condition.

Results and Discussion

Effect of solidifying agent on the compressive strength

The compressive strength of the solidification blocks treated by ISSA/PC and ISSA/PC/KL systems were shown in Figures 1 and 2. In ISSA/PC/KL system, incinerated sludge ash was mixed with fixed amount of PC (40 g/ 100g ISSA) and blended with different amounts of KL. The compressive strength of all solidified wastes incorporating the incinerated sludge ash was very low in the early age, and thereafter, the compressive strength increased to higher values gradually, as shown in Figure 1. This is because the calcium hydroxide crystal which was produced during the PC hydration process was consumed during the pozzolanic reaction of ISSA [17]. It also could be observed in Figure 1 that the more content of Portland cement in the solidified blocks, the higher compressive strength the solidified blocks show. The compressive strength of the solidification blocks increase little when the PC content of

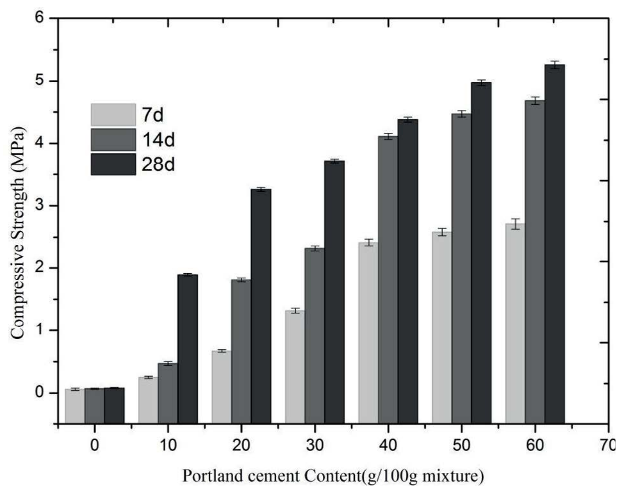

Figure 1: Effect of PC content on the compressive strength of solidification blocks under various curing time.

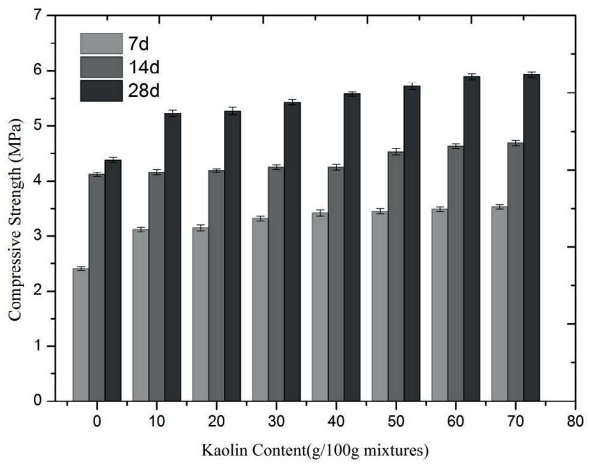

Figure 2: Effect of KLcontent on the **c**ompressive strength of solidification blocks under various curing time.

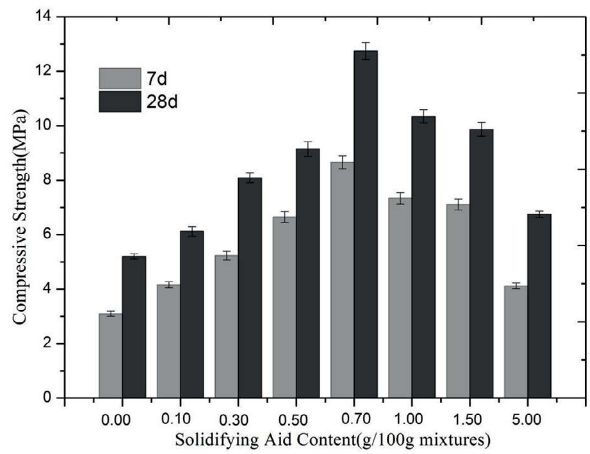

Figure 3: Effect of solidifying aid content on the compressive strength of solidification blocks.

the solidified agent exceeded 40g/100g mixture Figure 1. So, according to the cost of the solidification, 40g/100g mixture was chosen as the optimal content of PC. All the PC-solidified-samples met the minimum requirement for highway or the national highway subbase, but none met the minimum requirement for fired common bricks. It could be seen from Figure 2, when 10g/100g mixture of Kaolin was added to the system, the 28-day-compressive strength could develop from 4.38 MPa to 5.20 MPa. Thereafter, the compressive strength increased slowly with the increasing content of Kaolin. As a result, the 10g/100g mixture of Kaolin was chosen as the optimal content.

Effect of solidifying aid on the compressive strength

As shown in Figure 3, with different proportions of solidifying aid added to the solid mixtures including ISSA and solidifying agent with the fixed ratio mentioned in Table 3, the early-phase compressive strength of the samples was significantly improved compared to that of the samples without solidifying aid addition. It was probably because that the Na_2SiO_3-based solidifying aid accelerated the formation of ettringite, which has positive effect on the compressive strength of solidification block. The highest compressive strength of 12.74 MPa (shearing strength: 3.54 Mpa) was observed from solid mixtures blended with 0.70g/100g mixtures solidifying aid after 28 days curing, which meets the minimum requirement for fired common bricks. Subsequently, the compressive strength of the solidification blocks dropped when the content of the solidifying aid exceeded 0.70g/100g mixture. As a result, the 0.70 g/100g mixture was chosen as the optimal content of solidifying aid when the PC content was fixed at 40 g/100g mixture.

TCLP results

Leaching tests were conducted to examine the potential toxicity of heavy metal leaching from the solidified sludge ash, solidified block A (ISSA blended with PC and KL at the ratio of 100: 40: 10) and solidified block B (ISSA blended with PC, KL and SS-SA at the ratio of 100: 40: 10: 0.7). All the samples were cured under 20± 1°C for 28 days. The results of the metal concentrations in TCLP leachates were shown in Table 4. TCLP results showed that the concentrations of all the heavy metals in the leachate from solidified samples were all lower than the maximum

Component (mg/L)	ISSA	Block A	Block B	EPA hazardous Criteria
As	0.1217 ± 0.0002	0.0134 ± 0.0001	0.0073 ± 0.0001	<5.0
Ba	0.3352 ± 0.0003	0.2780 ± 0.0003	0.1592 ± 0.0002	<100
Cd	0.2930 ± 0.0003	0.2739 ± 0.0002	0.2672 ± 0.0002	<1.0
Cr	0.1367 ± 0.0002	0.0968 ± 0.0001	0.0887 ± 0.0001	<5.0
Ni	0.2170 ± 0.0002	0.0388 ± 0.0001	0.0331 ± 0.0001	<10.0
Pb	0.0122 ± 0.0001	ND	ND	<5.0

ND, not detected

Table 4: Toxic characteristics leaching procedure (TCLP) test results.

solubility limit set by the Environmental Protection Agency in the Republic of China, and thus the risk of hazardous heavy metals leaching following the use of solidified sewage sludge ash as construction materials may be safely concluded to be quite low. In addition, metal concentrations in the TCLP leachates extracted from the cement-based solidified ISSA were much lower than that extracted from the untreated ISSA. It could be concluded that the heavy metal could be immobilization by cement-based solidification, and the addition of solidifying aid could enhance the immobilization of heavy metals.

X-ray diffraction analysis

The crystalline phases presented in the solidified waste samples were characterized by X-ray diffraction (XRD) analysis. The samples were mixtures of ISSA, PC, KL and Solidifying aid at a mass proportion of 100:40:10:0.7, which was obtained from above research results. Samples cured at 20 ± 1°C for 28 days were crushed into powder using a mortar and pestle. Then the powder was sieved through a 150 μm mesh and scanned from 0° to 60°. The XRD analysis results were shown in Figure 4. As presented in Figure 4(a) and Figure 4(c), X-ray diffraction experiments provided variation information about the crystalline material among the different samples. The results of the XRD analysis revealed that the major crystalline phases presented in the Figures were quartz (SiO_2), CSAH ($CaO{\bullet}SiO_2{\bullet}Al_2O_3{\bullet}nH2O$), ettringite ($3CaO{\bullet}Al_2O_3{\bullet}3CaSO_4{\bullet}32H_2O$) and CSH ($CaO{\bullet}SiO_2{\bullet}nH_2O$). These hydrated compounds are generally formed by a Pozzolanic reaction through the reaction of $Ca(OH)_2$ and SiO_2 [18]. It could be observed from Figure 4(a) that, quartz, one of the main chemical components of ISSA, is prominent in the diffraction patterns of the untreated ISSA blocks. No other crystalline phases matched sufficient peaks to be positively identified, although analysis using an XRD pattern data base (International Centre for Diffraction Data, ICDD) suggested minor peaks may correspond to aluminium phosphate ($AlPO_4$), iron silicite ($FeSi_2$) and calcium copper fluoride ($CaCuF_4$) [19]. When ISSA was blended with solidifying agent at a ratio of 5:2 Figure 4(b), ettringite and CSH were both detected, which complied with relevant research reporting that the hydrated products of the cement were known to be CSH, CAH, CSAH, ettringite, C_3AF ($3CaO•Al_2O_3•Fe_2O_3$), $Ca(OH)_2$, $CaSO_4$ and $CaCO_3$ [20]. Similar XRD pattern was observed when solidifying aid was added to the ISSA/PC/KL system Figure 4(c).

Scanning electron microscopy analysis

Figures 5(a) and 5(b) showed the SEM observations of the incinerated sludge ash samples before and after solidification, respectively. Figure 5(a) was the SEM image of the unsolidified incinerated sludge ash and Figure 5(b) was the solidified block A, ISSA blended with PC and KL at the ratio of 100:40:10. As shown in the SEM micrographs, honeycomb-like hydrated products, known to be distinct CSH compounds [21] which have positive relationship with the compressive strength, were not observed in the incinerated sludge ash before solidification Figure 5(a). On the contrary, they were found to be widespread on the surface of the solidified sludge ash mixed with PC and KL.

Conclusion

The solidification of ISSA mixed with PC and KL was investigated to examine the feasibility of its utilization as construction materials with the purpose of the economical recycling of waste materials. Compressive strength tests have showed that incinerated sludge ash solidification using PC, KL and SS-SA in appropriately mixed proportions enhanced the geotechnical characteristics of the sludge ash, with an ultimate strength meeting the minimum requirement for fired common bricks. TCLP results showed that the metal concentrations in leachates extracted from the cement- based solidified ISSA were much lower than that from the untreated ISSA, far below the EPA (China) toxicity characteristic criteria, suggesting that the solidification can minimize the hazard of heavy metals leaching from the solidified ISSA. Quartz, CSAH, ettringite and CSH were found in the XRD patterns of the 28-day solidified samples, which play an important role in enhancing the solidifying reaction of the sludge ash. SEM micrographs showed that the structure compaction was due to the formation of honeycomb-like C-S-H compounds caused by the addition of solidifying agent. It could be concluded that solidification and recycling for construction materials could be an inexpensive and effective treatment method for incinerated sewage sludge ash.

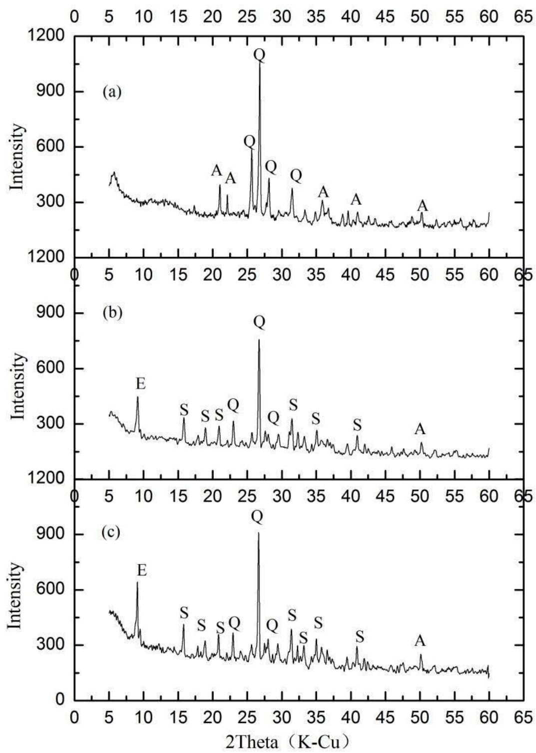

Figure 4: XRD patterns of three different samples. (a): untreated ISSA; (b): block A; (c): block B.

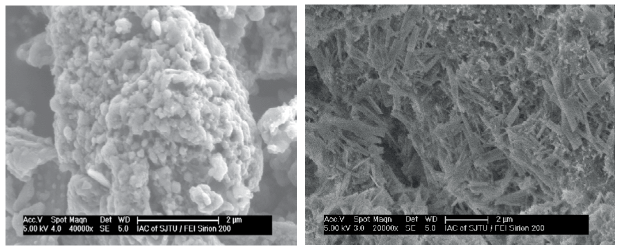

Figure 5: SEM Photographs of unsolidified ISSA and solidified block A. (a): unsolidified ISSA; (b): block A.

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