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Transformation of hazardous residues from laboratory waste treatment plant into compressed concrete paving units



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Abstract

In this study, we investigated the transformation of hazardous residues, including fly ash, silica gel, bottom ash, and sludge, from a laboratory waste incineration system at the Sustainable Environment Research Center into compressed concrete paving units (CCPU). The raw fly ash was washed twice with water to remove Cl⁻. The secondary washed fly ash (SWFA) was then mixed with silica gel, bottom ash, and sludge using various formulations designed through response surface methodology (RSM) in Design Expert 8.0.6 software. The mixed materials were subsequently melted at 1,400 °C. The results of the RSM analysis indicated that the optimal conditions were achieved when the encapsulating phase (silica gel) comprised 60% of the total sample, while the washed fly ash constituted 10% of the encapsulated phase (SWFA, bottom ash, and sludge). This configuration minimized the amount of unmelted materials and maximized the percentage of mass reduction. The slag with optimal melting properties was combined with cullet, clay, cement, and water, then molded, cured, and transformed into CCPU. The physical properties of specimens met the standards for Grade-A CCPU, and the leaching availability results indicated that metals were effectively encapsulated within the CCPU. Overall, the CCPU provides adequate physical strength and will not contribute to environmental pollution when recycled.

Keywords Fly ash, Slag, Response surface methodology, Compressed concrete paving units, Melting

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1 Introduction

In the fields of education and research, laboratory waste (LW) is inevitably generated during experiments. LW can be challenging to dispose of due to its diverse nature and small quantities. To tackle this issue, the Sustainable Environment Research Center (SERC) at National Cheng Kung University established a LW treatment plant in 2005. This facility features three distinct treatment systems: an incineration system, a physicochemical treatment system, and a plasma melting system. The input materials for this plant primarily originate from chemical and medical laboratory experiments. Among these, combustible medical laboratory waste and plastic containers from medical laboratories (as opposed to hospitals) are directed to the incineration system. Liquid LW is classified based on its characteristics, such as organic LW,



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acidic LW, or LW containing heavy metals, and is subsequently sent to various units within the physicochemical treatment system.

During the two treatment processes mentioned above, residues such as fly ash, bottom ash, and sludge are generated and sent to the plasma melting system. Plasma melting technology has been utilized to melt a wide variety of materials, including glass mixtures and solid waste incinerator fly ash [1, 2]. The mixture of residues was mixed with silica gel or cullet rich in SiO₂ and heated to 1,400 °C using a plasma torch. The high temperature decomposed the organic materials, while the SiO₂ served as a glassy matrix to encapsulate the hazardous substances [3]. The molten materials were then quenched with water to form glassy slag. Following the thermal melting process, the slag was transformed into an inert form, and the hazardous materials were either decomposed or stabilized. According to previous studies, the slag can be recycled as composite materials or building materials [4, 5]. However, the combustible medical LW from medical or biochemical experiments typically contains a high concentration of Cl⁻. The presence of Cl⁻ leads to the formation of polychlorinated dibenzop-dioxins and dibenzofurans (PCDD/Fs) and HCl during the incineration process [6, 7]. The HCl of the flue gas was neutralized by NaOH solution injected into the quenching tower, resulting in the formation of NaCl present in the fly ash [8]. Consequently, the fly ash contained extremely high levels of Cl⁻ and PCDD/Fs, necessitating treatment in a plasma melting furnace. During the melting process, Cl⁻ increased the required melting temperature and hindered the formation of slag. Therefore, it may be beneficial to remove Cl⁻ prior to melting to enhance stabilization. A washing process was proven to effectively eliminate Cl⁻, and this washing process also enhanced the melting effect [9]. After the melting process, the waste was converted into slag, and recycling this material presented another critical challenge. Various methods were employed to recycle stabilized waste, such as blast furnace slag, fly ash, or gangue, into building materials. To achieve adequate strength, heating or high pressure may be required during the manufacturing process [4, 10, 11].

In this study, the raw fly ash (RFA) from the incineration system was washed with water to remove Cl⁻. The washed fly ash was then mixed with silica gel (sourced from the LW treatment plant), bottom ash (collected from the incineration system), and sludge (sampled from the physicochemical treatment system) in various mass ratios. During the melting process, the silica gel served as the glassy matrix to encapsulate hazardous materials, functioning as the encapsulating phase. The other three materials, the fly ash, the bottom ash, and the sludge, were identified as hazardous materials to be encapsulated within the glassy matrix, referred to as the encapsulated phase. The mixtures were melted at 1,400 $^{\circ}$ C in a laboratory-scale heating furnace and subsequently aircooled without any ventilation to form slag. The composition and leaching of metals in the slags were examined to assess the effectiveness of vitrification. The slag demonstrating the most effective vitrification was reused to create compressed concrete paving units (CCPU). The chemical properties and physical strength of the CCPU were evaluated to determine the feasibility of recycling slag as a building material.

2 Experimental sections

2.1 Preparation of raw materials

The flowchart of the LW incineration system at SERC was presented in our previous study [12]. The fly ash and bottom ash were collected from the primary quenching tower and the bottom ash pit of the incineration system, respectively. The sludge samples were taken from the filter press plate of the heavy-metal-based liquid LW treatment unit. The silica gel and cullet were obtained from the temporary storage area of the LW treatment plant. All the specimens were dried and ground to a powder with a particle size smaller than 150 μ m (ASTM 100 mesh) to ensure their uniformity for subsequent experiments.

2.2 Procedure of the washing, melting, and CCPU making processes

Figure 1 illustrates the process flow chart of the experiments. The RFA was mixed with deionized water in a mass ratio of 1:5. The mixture was stirred for 10 min and then filtered using a mixed cellulose ester filter with an average pore size of 1 µm to remove suspended solids. The first washed fly ash (FWFA) was subsequently dried, weighed, and analyzed. The FWFA then underwent the same washing, filtration, drying, and analysis process as the second washed fly ash (SWFA). Additionally, the wastewater from the first and second washing processes was filtered for further analysis. The melting behavior of various materials was evaluated using RSM, which was previously employed to investigate the melting behavior of a fluidized bed [13]. The mixing formula for the melted materials (SWFA, silica gel, bottom ash, and sludge) was developed using Design Expert 8.0.6 software (Stat-Ease Inc.). To assess how the mixing formula of these waste materials influenced the melting processes, we utilized two expected yields as response variables for the control factors. The first yield represented the remaining mass of input materials that was not converted into slag (referred to as unmelted materials). The second yield indicated the reduction mass percentage of the input materials during the thermal melting process.



Among the melted materials, silica gel was identified as the encapsulating phase, while the SWFA, bottom ash, and sludge comprised the encapsulated phase. Two operational parameters were utilized in the experiments: the mass fraction of the encapsulating phase and the mass fraction of SWFA. The mass fraction of the encapsulating phase was defined to range from 50 to 70%, while the mass fraction of SWFA was set between 5 and 15%. The mass ratio of sludge to bottom ash was set at 5:3. According to the initial design, a total of 13 sets of test runs were planned for the experiment. The analyzed data was subsequently processed and imported into the software. By plotting a contour graph on a plane with independent variables as axes, connecting points with the same function value produced contour lines. These contour lines illustrated the relationship between the response and independent variables, creating a contour plot of a three-dimensional surface. This contour plot aided in analyzing the optimal operating conditions for this research. The total mass of the raw materials was 40 g, and the formulas for the 13 test runs are presented in Table 1. The basicity (mass of CaO/mass of SiO_2) of the raw materials, calculated based on the elemental composition results, was also shown in Table 1. Due to the difficulty in melting, the proportion of the encapsulating phase must be higher, ranging from 50 to 70%. Our previous research indicated that the fly ash of this incinerator was the material least likely to melt, so its mixing ratio was set between 1.5 and 7.5% [14]. The mixing ratios for sludge and bottom ash were set to range from 15.9 to 29.7% and 9.6 to 17.8%, respectively. The specimens were heled in graphite crucibles and heated using an electric furnace. The operating voltage was 220 V, and the maximum operating temperature reached 1,500 °C. The heating program was as follows: from room temperature to 800 °C at a rate of 10 °C min⁻¹ from 800 to 1,200 °C at 6 °C min⁻¹, from 1,200 to 1,400 °C at 4 °C min⁻¹, and held at 1,400 °C for 1 h. The melted specimens were then cooled to room temperature without forced ventilation, resulting in the formation of slags. Each

Run	Encapsulating phase	Encapsulated ph	Encapsulated phase				
	Silica gel (%)	SWFA (%)	Sludge (%)	Bottom ash (%)			
R1	60	4	22.5	13.5	0.19		
R2	70	1.5	17.8	10.7	0.17		
R3	60	4	22.5	13.5	0.19		
R4	50	5	28.1	16.9	0.21		
R5	50	7.5	26.6	15.9	0.20		
R6	60	2	23.8	14.2	0.19		
R7	70	4.5	15.9	9.6	0.17		
R8	60	6	21.2	12.8	0.18		
R9	50	2.5	29.7	17.8	0.21		
R10	60	4	22.5	13.5	0.19		
R11	70	3	16.9	10.1	0.17		
R12	60	4	22.5	13.5	0.19		
R13	60	4	22.5	13.5	0.19		

slag was labeled with an S-number corresponding to the experimental batch number.

The slags were pulverized and analyzed for their physical and chemical properties to evaluate the impact of the thermal melting process. Previous studies have demonstrated that the recovered materials, such as fly ash or slag, can be converted into concrete blocks through compression and sintering [10, 11]. In this study, the slag was pulverized into a fine powder and then mixed with cullet, clay, cement, and water according to the formulas presented in Table 2. Two rounds of CCPU-making tests were conducted. Each CCPU was assigned a unique CCPU number corresponding to the experimental batch number it belonged to. A previous study indicated that the curing process improved physical strength [15]. Consequently, the CCPU were cured for 28 d, and their compressive strength was measured at various intervals, ranging from 7 to 28 d, to evaluate their physical properties.

2.3 Analysis of element composition and metal leaching behavior

To analyze the metal composition of solid specimens, a procedure modified from the standard method outlined in NIEA R317.12 C was conducted [16]. The solid specimens were pulverized and ground to a particle size smaller than 150 μ m (ASTM 100 mesh) to ensure their uniformity. Approximately 0.1 g of the ground samples was mixed with a concentrated acid mixture consisting of 3 mL HCl (32%), 5 mL HNO₃ (67%), and 0.5 mL HF (32%), and then placed in sealed Teflon tubes. These tubes were heated using a microwave digester (MARS Xpress, CEM) according to the following temperature control program: heating from room temperature to 180 °C at a rate of 5 °C min⁻¹, from 180 to 210 °C at a rate of 3 °C min⁻¹, held isothermally for 15 min, and finally cooling down to room temperature with forced ventilation. The digested solutions were diluted with deionized water to a final volume of 50 mL and filtered using mixed cellulose ester filters to remove suspended solids for further analysis.

The characteristic leaching procedure for building materials was employed to evaluate the leaching availability of inorganic components in the CCPU [17]. The CCPU specimens were ground to a particle size of less than 125 μ m (ASTM 120 mesh). The deionized water was mixed with the specimens at a liquid-to-solid ratio (L S⁻¹) of 50 L kg⁻¹ for the two-stage extraction process. In the first

Table 2 Formula of raw materials of CCPU (in%)

Run	Cullet	Clay	Cement	Slag	Water
1	13.3	27.5	15	13.2	30
2	0	27.5	15	27.5	30

stage extraction, 10 g of the specimen (denoted as M in Eqs. (1) and (2)) was mixed with 500 mL of deionized water (denoted as V_0 in Eq. (2)) and stirred. The pH of the leaching solution was adjusted to 7 by adding 1 M-HNO₃ solution (denoted as V_1 in Eqs. (1) and (2)) if the pH of mixture was higher than 7. After stirring for 3 h, the extract was collected. In the second stage extraction, the specimens were mixed again with deionized water, and the pH of the leaching solution was adjusted to 4 by adding 1 M-HNO₃ solution (denoted as V_2 in Eqs. (1) and (2)) if the pH of mixture was higher than 4. After stirring for an additional 3 h, this extract was also collected. The extracts from both stage extractions were mixed, and the concentrations of the resulting mixture were analyzed to determine the leaching availability of inorganic substances and the acid neutralizing capacity (ANC, in mol kg^{-1}).

$$ANC = \frac{V_1 + V_2}{M(1 - \theta)}$$
(1)

 V_1 = the volume of 1 M-HNO₃ solution added in the first stage extraction (mL).

 V_2 = the volume of 1 M-HNO₃ solution added in the second stage extraction (mL).

M = the mass of specimen (g).

 θ = water content (%).

The concentrations (denoted as C in the following equation) of Al, Ca, Cd, Co, Cr, Cu, Fe, K, Mn, Mg, Na, Ni, Pb, Si, and Zn in the digested solutions, washing wastewater, and extracts were measured using atomic absorption spectroscopy (Agilent Technologies 55 AA). The availability of metals ($U_{availability}$, in mg kg⁻¹) was calculated as Eq. (2).

$$U_{\text{availability}} = \frac{C(2V_0 + V_1 + V_2)}{1000M(1 - \theta)}$$
(2)

C = the metal concentration of leaching solution (mg L^{-1}).

 V_0 = the volume of deionized water added in the first and second stage exactions (mL).

 V_0 (mL), V_1 (mL), V_2 (mL), M (g), and θ (%) are the same as Eq. (1)

The detail procedures for preparing the leaching solution, determining the leaching parameters, filtering the mixture, and calculating availability and ANC all adhered to the Standard Methods outlined in the NIEA R218.10 C [17].

2.4 Evaluation of physical property of solid specimens

The solid specimens were pulverized and ground to a particle size smaller than 150 μ m (ASTM 100 mesh) to ensure their uniformity. The crystalline phases in the slags were determined using X-ray diffraction (XRD)

analysis. This analysis was conducted with a powder diffractometer (Geigerflex 3063) utilizing Ni-filtered Cu Ka radiation on powders with a particle size smaller than 20 μ m, at a scanning rate of 4° min⁻¹, in the 2 θ range of 10-60°. Crystalline phases were identified by comparing the intensities and positions of Bragg peaks with those listed in the Joint Committee on Powder Diffraction Standards data files. Specimens pulverized to particles smaller than 74 µm (ASTM 200 mesh) were affixed to a metallic plate, coated with a gold film, and qualitatively examined for the microstructure of the slags using scanning electron microscopy (SEM, Jeol JXA- 840). The compressive strength of the CCPU was measured following the standard method outlined in Chinese National Standards (CNS) 13295 [18]. The CCPU specimens were prepared in dimensions of 5 cm \times 5 cm \times 5 cm. The load was increased at a rate of 0.2-0.3 N mm² s⁻² until the load at which the specimen ruptured was reached. The compressive strength was then calculated by dividing the load at rupture by the cross-sectional area of the specimen.

3 Results and discussions

3.1 Metal migration of fly ashes during the washing processes

Table 3 presents the metal composition of the RFA, FWFA and SWFA. The predominant elements of RFA were Na (133 g kg⁻¹) and Cl⁻¹ (112 g kg⁻¹), followed

by Fe (25.4 g kg⁻¹) and Si at (22.4 g kg⁻¹). The levels of other metals were all below 10.0 g kg⁻¹. Following the first and second washing processes, the remaining mass percentages of FWFA and SWFA decreased to 81 and 72%, respectively. During the washing processes, watersoluble ions were removed, resulting in a significant reduction in the content of most elements. The most notable decreases were observed in the Na and Cl⁻ levels. The levels of Na and Cl⁻ in the fly ash decreased from 133 g kg⁻¹ and 112 g kg⁻¹ to 4.2 g kg⁻¹ and 8.5 g kg⁻¹, respectively.

Table 4 displays the mass fraction of elements in washed fly ashes and wastewater during the two washing processes. Among these elements, those with a total washing percentage exceeding 70% included Ca (78.2%), Co (75.6%), K (94.7%), Mg (83.6%), Na (97.2%), Ni (81.6%), Zn (72.7%), and Cl⁻ (93.1%). These components primarily remained in the washing wastewater, significantly reducing the mass of fly ash by approximately 30% during the two-stage washing process. The washing wastewater contained a substantial amount of metals and cannot be discharged directly. For the practical application of SERC, the washing wastewater must be pumped to a physicochemical treatment system for further processing. Additionally, the secondary sludge generated in the physicochemical treatment system requires thermal melting in the plasma melting furnace.

Item	RFA		FWFA		SWFA		
	Average (g kg ⁻¹)	RSD (%)	Average (g kg ⁻¹)	RSD (%)	Average (g kg ⁻¹)	RSD (%)	
Al	1.3	57.1	1.1	45.4	0.6	41.1	
Ca	6.9	67.0	3.4	16.8	1.7	8.5	
Cd	0.04	2.4	0.04	15.0	0.04	3.4	
Со	0.06	4.4	0.03	27.7	0.02	2.3	
Cr	4.8	18.5	5.2	51.1	3.5	27.0	
Cu	1.3	12.3	0.9	12.2	0.7	3.5	
Fe	25.4	21.8	20.8	17.6	24.9	33.3	
К	2.1	57.0	0.5	31.1	0.1	12.8	
Mn	0.3	7.9	0.2	27.6	0.09	4.0	
Mg	1.9	4.5	1.2	43.9	0.3	12.4	
Na	133	1.3	47.4	48.7	4.2	5.3	
Ni	1.6	6.3	0.8	39.5	0.3	34.8	
Pb	3.2	23.4	2.1	3.87	2.2	6.4	
Si	22.4	21.1	17.5	14.2	14.5	15.3	
Zn	1.8	2.2	1.1	40.9	0.5	4.7	
CI ⁻	112	19.4	36.5	13.5	8.5	16.2	
RMP (%)	100	-	80.7	12.3	72.1	14.5	

RMP (Remained mass percentage, %) = $\frac{\text{remained mass of washed fly ash}}{\text{mass of RFA}} \div \times 100\%$

ltem	First washing process		Secono proces	lary washing s	Total washing percentage	
	FWFA	Wastewater	SWFA	Wastewater	(%)	
AI	61.8	38.2	71.0	29.0	59.9	
Ca	27.6	72.4	45.0	55.0	78.2	
Cd	96.1	3.9	95.0	5.0	8.6	
Co	40.2	59.8	64.9	35.1	75.6	
Cr	77.1	22.9	90.3	9.7	33.6	
Cu	51.1	48.9	60.9	39.1	52.7	
Fe	83.3	16.7	94.0	6.0	11.8	
К	14.2	85.8	24.1	75.9	94.7	
Mn	36.6	63.4	49.5	50.5	70.9	
Mg	35.3	64.7	51.2	48.8	83.6	
Na	23.0	77.0	12.5	87.5	97.2	
Ni	29.3	70.7	2.4	99.9	81.2	
Pb	80.7	19.3	95.9	4.1	38.6	
Si	46.8	53.2	83.4	16.6	41.7	
Zn	47.9	52.1	40.0	60.0	72.7	
CI-	21.2	78.8	30.5	69.5	93.1	

 Table 4
 Washing behavior of the two washing processes

Figures 2 and 3 show the XRD analysis results and the SEM images of the fly ash and the washed ashes. In Fig. 2a, NaCl was the main crystalline phase of RFA, and its intensity significantly surpassing that of CaCO₃. The presence of NaCl resulted from the reaction between the injected NaOH solution and HCl gas of the flue gas, corroborating findings from a previous study [8]. Following the first washing process, the intensity of NaCl was markedly reduced, while the baseline intensity increased significantly. After the second washing process, the NaCl crystals were no longer detectable in the XRD diagram. The XRD analysis results indicated that a significant amount of NaCl was removed from the fly ash during the washing process, which is consistent with the SEM findings presented in Table 3.

In Fig. 3a, numerous porous crystals were observed on the surface of RFA. According to our previous study, these porous crystals were confirmed to be NaCl [8]. As the water washing process continued (as in Figs. 3b and 3c), the quantity of porous crystals gradually decreased, and a block-like structure began to emerge. Both the XRD analysis results and the SEM images were consistent with the elemental content analysis results presented in Table 3.

3.2 Evaluation of melting effect

Table 5 shows the metal composition of the input materials used in the thermal melting process. The SWFA contained Fe (24.9 g kg⁻¹), Si (14.5 g kg⁻¹), Cl (8.5 g kg⁻¹), Na (4.2 mg kg⁻¹), and along with various trace





Fig. 2 XRD analysis result of fly ash and washed fly ashes: a RFA; b FWFA; c SWFA

elements. The silica gel was primarily composed of Si (92.5 g kg⁻¹) with trace amounts of other elements. The sludge predominantly consisted of Fe (15.2 g kg⁻¹), Na (39.1 g kg⁻¹), Ca (9.5 g kg⁻¹), Cu (24.1 g kg⁻¹), and Si (15.2 g kg⁻¹). The bottom ash mainly contained mainly Fe (70.8 g kg⁻¹), Si (37.8 g kg⁻¹), Al (31.3 g kg⁻¹), Na (22.5 g kg⁻¹), and Ca (36.5 g kg⁻¹). Based on the elemental composition of the input material, the basicity of the mixture was calculated, as shown in Table 1. According to the previous study, the fly ash from the incinerator at the SERC was difficult to melt [14]. Consequently, the basicity of the input material mixture was set between 0.17 and 0.21, which was significantly



Fig. 3 SEM images of fly ash and washed fly ashes: a RFA; b FWFA; c SWFA

lower than the typical basicity used in the melting process [19].

Figure 4 illustrates the mass percentage of unmelted materials across various mixing formulas. The highest proportion of unmelted slag, recorded at 1.8%, was observed when the content of SWFA in the encapsulated material was increased, while the silica gel content was decreased. In other test processes, the proportion of unmelted materials varied from 0.21 to 0.87%. Conversely, an increase in the proportion of silica gel led to a

decrease in the amount of unmelted materials during the melting process.

Figure 5 illustrates the percentage of mass reduction with different mixing formulas during the melting process. The experimental results indicated that as the proportion of SWFA and silica gel additives increased, the mass reduction percentage also increased. The intricate composition of SWFA was the primary reason for this, as when heated to 1,400 °C during the melting process, volatile elements (mainly chloride) evaporated into the flue gas. Moreover, a higher proportion of silica gel additives decreased the basicity to enhance the treatment effect on the melted materials and similar results were also reported in other study [20]. However, adding more silica gel did not necessarily lead to better effects. Overall, the experimental results of the RSM indicated that the most optimal effect was achieved when the silica gel constituted 60% of the total sample and the SWFA constituted 10% of the encapsulated phase by mass, in terms of minimizing the amount of unmelted materials and maximizing the percentage of mass reduction.

Table 6 presents the slag compositions under three different conditions. The elemental compositions of these slags were generally similar, with the primary difference being the Si content. In the case of S- 5, the raw materials included the maximum amount of SWFA (7.5%) and the minimum quantity of silica gel at 50%. The mixing formula resulted in a basicity of 0.20 for run- 5, which was relatively high among these tests and led to a decrease in Si content (42.7 g kg⁻¹). Consequently, the mass percentage of unformed material was relatively high.

Conversely, the formula for run- 7 contained the highest proportion of silica gel (70%) and a moderate amount of SWFA (4.5%). The basicity of run-7 was reduced to 0.17, resulting in a significant increase in Si content (108 $g kg^{-1}$). The increase in the amount of silica gel contributed to more formation. However, when comparing S-3 and S-7, S-3 exhibited a lower mass fraction of the encapsulating phase (60%) and a relatively low mass percentage of unformed material. S- 3 was identified as the slag with the most optimal thermal melting effect; consequently, the potential for metals to vaporize into the flue gas in this run was also discussed (as shown in Table 6). The approximate gas phase fraction (%) was defined as (1- metal mass of slag/metal mass of raw materials) $\times 100\%$. The results show that most elements tended to remain in the slag after the melting process, except for Cd, Pb, Zn and Cl⁻, which had an approximate gas phase fraction greater than 90%. According to the previous study, Cd, Pb, and Zn had low boiling points and primarily existed in particulate form [21]. Under typical high temperature conditions, Cl⁻ was transformed into HCl, which could also lead to the formation of PCDD/Fs [6,

ltem	SWFA	SWFA		Silica gel			Bottom ash	
	Average (g kg ⁻¹)	RSD (%)	Average (g kg ⁻¹)	RSD (%)	Average (g kg ⁻¹)	RSD (%)	Average (g kg ⁻¹)	RSD (%)
Al	0.6	41.1	N.D	N.A	8.7	2.8	31.3	9.2
Ca	1.7	8.5	11.2	38.2	9.5	8.6	36.5	32.8
Cd	0.04	3.4	N.D	N.A	0.4	3.3	< 0.001	49.5
Со	0.02	2.3	N.D	N.A	0.7	4.7	0.5	3.7
Cr	3.5	27.0	0.01	69.6	0.6	5.0	2.8	8.4
Cu	0.7	3.5	N.D	N.A	24.1	2.96	7.1	7.5
Fe	24.9	33.3	0.4	15.9	15.2	10.0	70.8	7.2
К	0.1	12.8	0.02	14.0	6.4	7.4	2.7	8.7
Mg	0.3	12.4	3.6	8.1	1.0	11.6	9.0	18.9
Mn	0.09	4.0	N.D	N.A	1.6	4.3	2.2	26.4
Na	4.2	5.3	1.9	7.2	39.1	8.0	22.5	20.3
Ni	0.3	34.8	N.D	N.A	5.2	4.6	2.0	5.3
Pb	2.2	6.4	N.D	N.A	3.2	3.8	1.5	5.6
Si	14.5	15.3	92.5	21.8	15.2	31.1	37.8	28.4
Zn	0.5	4.7	0.05	17.8	1.0	3.8	2.5	5.3
CI-	8.5	16.2	0.8	15.4	9.4	28.5	1.4	25.4

Table 5 Element content of input-material of thermal melting process

N.D. Not Detected, N.A. Not Available

22]. However, the temperature during the melting process reached up to 1,400 °C and such high temperatures would decompose organic substances, resulting in only trace amounts of PCDD/Fs remaining in the particulate phase [14]. Therefore, air pollution control devices capable of removing particulates and HCl(g) are necessary for practical applications in industrial plants.

3.3 Transformation of slags into CCPU

Due to the optimal effects of thermal melting, S- 3 was selected as the raw material for CCPU. Table 7 presents the elemental composition of the raw materials used in the production of CCPU. The cullet primarily consisted of Si (376 g kg⁻¹), with additional amounts of Al (54.1 g kg⁻¹) and Ca (29.0 g kg⁻¹). The clay was predominantly composed of Si (185 g kg⁻¹), Al (92.6 g kg⁻¹), and Fe (17.8 g kg⁻¹). The elemental composition of the cement was mainly Ca (282 g kg⁻¹) and Si (254 g kg⁻¹). The mixed raw materials with two different formulations are mixed with water, molded, cured, and subsequently transformed into CCPU.

Figure 6 illustrates the compressive strength of CCPUs over various curing days. The compressive strength of CCPU- 1, and CCPU- 2 increased gradually throughout the curing period. The CCPU- 2 exhibited a strength increase of approximately 5% compared to the CCPU- 1. The physical strength of both CCPU- 2 and CCPU- 1 met the CNS 13295 Grade A CCPU criteria on the 21 st and 28 th curing days [18]. The compressive strength of the

CCPUs was approximately equal to that of bricks produced under a molding pressure of 50 kg cm⁻² at a concrete-brick manufacturing plant; however, it was weaker than that of general high-pressure bricks and calcium carbonate high-pressure bricks [23]. This discrepancy may be attributed to the relatively small quantity of concrete used and the reduced pressure applied during the concrete-brick manufacturing process. When comparing CCPU- 1 and CCPU- 2, substituting cullet for slag did not enhance the physical strength of CCPU. This could be due to slag possessing superior physical properties compared to cullet, making it more suitable for blending with clay and cement. Other CCPU produced using desulfurization slag also demonstrated superior physical properties, as reported in a previous study [24].

Table 8 displays the leaching availability and the leaching mass fraction of inorganic component of CCPUs. The leaching mass fraction of inorganic component (%) was defined as $[U_{availability} (mg kg^{-1})/$ Composition of element $(mg kg^{-1})] \times 100\%$. The leaching availability of hazardous metals in CCPU- 1 and CCPU- 2 was as follows: Cd (ND, ND), Cr (0.15 mg kg^{-1}, 0.24 mg kg^{-1}), Cu (0.28 mg kg^{-1}, 0.35 mg kg^{-1}), Ni (0.13 mg kg^{-1}, 0.08 mg kg^{-1}), and Zn (0.75 mg kg^{-1}, 0.08 mg kg^{-1}). For the hazardous metals, the leaching availability values were below 1 mg kg^{-1}) and the leaching mass fractions were all less than 0.1%. For the crustal metals, the major metal species leached out were Al, Ca, Fe, and Mg and the leaching mass



Fig. 4 Mass percentage of unmelted materials for different operation parameters

fractions were less than 1%. Additionally, the ANC of CCPU- 1 and CCPU- 2 was 1.93 and 1.85 mol kg⁻¹, indicating that 1 kg of CCPU could neutralize approximately 2 mol of acid. The leaching availability results suggested that metals were effectively encapsulated in the CCPU, which demonstrated an acid neutralization ability of about 2 mol. Therefore, the chemical property results indicated that recycling slag into the CCPU could resist acid corrosion and would not lead to secondary environmental pollution.

3.4 Crystalline characteristics and surface structure of slags and CCPU

Figure 7 shows the SEM images of various slags. In Fig. 7b, numerous fine powders were visible on the surface of S- 5, which exhibited a porous structure. This porosity was attributed to the excessive addition of SWFA, leading to in a high Cl content. The vaporization of Cl^- contributed to the formation of the porous structure and hindered the vitrification of the mixture. In Fig. 7a, the small powders in S- 3 was not found,



Fig. 5 Percentage of mass reduction during melting process for different operation parameters

revealing a glassy and amorphous structure. This change could be explained by the increased addition of silica gel and the reduced addition of SWFA, which enhanced the effect of vitrification. Figure 7c shows that the structure of S- 7 was similar to that of S- 3, although some fine powdery grains were present on the amorphous structure. Compared to the input materials in run- 3, those in run- 7 contained a higher proportion of silica gel and SWFA. Due to the partial volatilization of silica gel and SWFA, run- 7 exhibited the highest mass reduction among all the slags.

Figure 8 shows the XRD analysis of the slags. The primary crystalline phase identified in S- 3, S- 5, and S- 7 was SiO₂, which resulted from the melting and reformation of silica gel. Additionally, Fe₂O₃ was detected in all three slags. In Fig. 8b, the baseline intensity of S- 5 was notably higher than that of S- 3 and S- 7, suggesting that the quantity of SiO₂ in S- 5 was lower than in S- 3 and S- 7. This discrepancy explained a poorly formed glassy matrix in S- 5, leading to an increase in unmelted materials, which corroborated the results of the RSM. In the full-scale plasma melting furnace, the input materials

ltem	S- 3			S- 5		S- 7	
	Average (g kg ⁻¹)	RSD (%)	Approximate gas phase fraction (%)	Average (g kg ⁻¹)	RSD (%)	Average (g kg ⁻¹)	RSD (%)
Al	7.6	5.8	13.7	8.9	18.9	5.4	8.0
Ca	18.8	2.5	5.0	16.7	7.73	12.6	11.4
Cd	N.D	N.A	> 99.9	N.D	N.A	N.D	N.A
Со	0.3	5.8	14.1	0.4	4.79	0.2	7.1
Cr	2.5	6.0	4.1	4.4	3.11	1.6	7.7
Cu	8.6	2.1	5.8	11.8	5.52	6.0	5.3
Fe	18.8	6.9	7.2	26.3	6.18	16.5	5.5
К	2.0	4.4	24.7	2.5	9.8	1.5	1.4
Mg	4.8	4.2	6.8	2.9	1.75	4.0	13.3
Mn	0.8	4.4	22.4	0.9	7.25	0.5	7.3
Na	12.4	15.2	33.9	19.2	10.3	16.7	8.7
Ni	2.0	2.4	6.6	2.7	2.81	1.6	5.8
Pb	N.D	N.A	> 99.9	N.D	N.A	N.D	N.A
Si	84.7	9.0	8.2	42.7	12.1	108	10.8
Zn	0.05	2.5	94.8	0.04	2.2	0.02	7.4
CI-	0.5	-	89.0	0.4	8.5	0.5	9.2
Basicity	0.19	-	-	0.20	-	0.17	-

Table 6 Element composition of slags and approximate gas phase fraction of S-3

N.D. Not Detected, N.A. Not Available

Table 7 E	lement content of	raw materia	is of CCPU
iable / E	lement content of	raw materia	IS OF CCPU

ltem	Cullet		Clay		Cement		S- 3	
	Average (g kg ⁻¹)	RSD (%)						
AI	54.1	3.7	92.6	22.8	7.5	23.1	7.6	5.8
Ca	29.0	4.7	1.7	8.0	282	16.6	18.8	2.5
Cd	N.D	N.A	0.01	16.3	N.D	N.A	N.D	N.A
Со	0.01	37.1	0.02	12.9	0.01	13.9	0.3	5.8
Cr	0.02	43.9	0.3	21.4	0.08	12.4	2.5	6.0
Cu	0.07	12.6	3.9	8.6	0.01	24.7	8.6	2.1
Fe	0.6	29.3	17.8	10.7	19.1	5.9	18.8	6.9
К	0.08	8.6	0.01	74.2	1.8	8.4	2.0	4.4
Mn	N.D	N.A	0.6	17.1	0.4	3.0	4.8	4.2
Mg	6.3	7.2	0.5	56.2	29.8	15.5	0.8	4.4
Na	2.2	4.0	1.2	13.3	3.5	3.7	12.4	15.2
Ni	N.D	N.A	0.4	15.1	0.03	2.8	2.0	2.4
Pb	N.D	N.A	0.3	7.7	0.06	11.7	N.D	N.A
Si	376	36.4	185	25.8	254	32.1	84.7	6.5
Zn	0.1	29.3	3.5	2.3	0.06	15.6	0.05	2.5
CI-	0.8	19.6	0.3	31.5	0.02	38.5	0.5	8.3

N.D. Not Detected, N.A. Not Available

included incinerator fly ash, incinerator bottom ash, sludge, and silica gel, similar to those used in this study; however, the resulting slag was completely amorphous. This was because that the melting mixture was quenched in the full-scale plasma melting furnace [25]. In this study, the mixture was air-cooled, leading to the formation of a crystalline phase that differed from that in the full-scale plant.



Fig. 6 Compressive strength of CCPUs at different curing day

Table 8 Leaching availability of inorganic component and ANC of CCPU

ltem	CCPU- 1		CCPU- 2		
	Leaching Availability (mg kg ⁻¹)	Leaching mass fraction (%)	Leaching Availability (mg kg ⁻¹)	Leaching mass fraction (%)	
Al	351	0.71	243	0.59	
Ca	410	0.59	325	0.48	
Cd	ND	< 0.01	ND	< 0.01	
Co	0.151	0.24	0.23	0.19	
Cr	0.07	< 0.01	0.08	< 0.01	
Cu	0.28	< 0.01	0.35	< 0.01	
Fe	211	0.85	230	0.59	
К	2.8	0.36	3.1	0.27	
Mn	0.25	0.05	0.53	0.03	
Mg	32.4	0.41	28.5	0.41	
Na	18.5	0.46	23.4	0.38	
Ni	0.13	0.03	0.08	< 0.01	
Pb	0.05	0.04	0.07	0.05	
Si	20.5	< 0.01	11.8	< 0.01	
Zn	0.75	0.04	0.95	0.07	
ANC (mol kg ⁻¹)	1.93		1.85		

N.D. Not Detected, N.A. Not Available

4 Conclusions

The primary composition of RFA consisted of Na at 133 g kg⁻¹ and Cl⁻ at 112 mg kg⁻¹, both of which were largely removed during the two-stage washing process, resulting in reduced levels of Na (4.2 g kg⁻¹) and Cl⁻ (8.5 g kg⁻¹). By mixing the sludge, bottom ash, and silica gel using various formulations, the mixture was melted at 1,400 °C. The findings indicated that the amount of unmelted slag peaked at 1.8% when the

proportion of SWFA in the encapsulated material was higher and the amount of silica gel was lower. Furthermore, the presence of silica gel decreased the quantity of unmelted materials during the melting process. The inclusion of SWFA and silica gel increased the percentage of mass reduction. The addition of silica gel improved the melting process; however, increasing its quantity did not consistently produce better results. According to the RSM experimental data, the most





Fig. 8 XRD analysis results of slags: a S- 3; b S- 5; c S- 7

Fig. 7 SEM images of slags: **a** S- 3; **b** S- 5; **c** S- 7

optimal results were achieved when silica gel constituted 60% of the total sample and SWFA accounted for 10% of the encapsulated phase mass. The S- 3 formulation exhibited the best melting effect and was subsequently transformed into the CCPU after the mixing, molding, and curing processes. The physical strength of these CCPUs met CNS 13295 Grade-A standards after a 28-d curing period. The leaching availability results indicated that the leaching mass fractions of crustal metals (Al, Ca, Mg, and Si) were lower than 1%, while those of hazardous metals (Cd, Cr, Cu, Ni, Pb, and Zn) were 0.1%. These results demonstrated that the CCPU possesses satisfactory physical strength and would not pose a risk of environmental pollution when recycled.

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Authors' contributions

HHW and KLH conceptualized this study; LCC and YMK conducted investigation; LCC, and SWH performed formal analysis; YMK undertook visualization and methodology, and wrote the first draft; HHW and KLH reviewed and edited the article. All authors have read and agreed to the published version of the manuscript.

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Data availability

All data generated or analyzed during this study are contained within the article.

Declarations

Competing interests

The authors declare they have no competing interests.

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