Preparation of Glass-Ceramics via Cosintering and Solidification of Hazardous Waste Incineration Residue and Chromium-Containing Sludge

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ABSTRACT: Residues from the incineration of hazardous wastes are classified as hazardous byproducts because they contain heavy metals. Chromium-containing sludge (CCS) is industrial sludge produced during the electroplating process and includes heavy metals, such as Cr, Pb, and Cu. These heavy metals can infiltrate natural ecosystems and cause significant environmental damage. To limit the toxicity of leached products, hazardous waste incineration residues (HWIRs) can be repurposed as raw materials for producing glass-ceramics. In this study, we designed an orthogonal experiment to optimize the heat treatment process, yielding glass-ceramics with excellent properties and realizing heavy metal solidification. The toxic characteristic leaching procedure was used to determine the leaching toxicity of the cosintered solidified heavy metals, revealing that their solidification efficiencies exceed 90%. Moreover, X-ray diffraction analysis indicates that certain heavy metals participate in the formation of heavy-metal-containing crystal lattices (FeCr$_2$O$_4$ and PbFe$_{12}$O$_{19}$), thereby reducing their leaching concentration. These results show that cosintering HWIR and CCS is an effective approach for heavy metal solidification and provides valuable insights into its utilization for producing building materials.

1. INTRODUCTION

Incineration is one of the foremost methods for disposing of hazardous waste owing to its benefits, such as chemical reduction and resource utilization. However, it also produces hazardous waste incineration residue (HWIR), which is classified as HW18 on China’s hazardous waste list and contains heavy metal contaminants, such as Cu, Cd, Pb, and Zn, which have a high migration capacity. Without appropriate treatment, HWIR poses a significant threat to ecosystems and human health. Therefore, a suitable disposal method is required.

Chromium-containing sludge (CCS) is industrial sludge comprising multiple metals and a small amount of organic matter. The most abundant elements in CCS are Fe, Pb, Cr, and Cu. The combination of heavy metals and organic compounds in sludge complicates its treatment and disposal. The most common disposal methods include landfill, heavy metal recovery, thermochemical technology, and solidification/stabilization. These methods are associated with several problems, such as inefficient land utilization, a high volume of secondary pollution, poor long-term stability of solidified heavy metal and organic byproducts, and high energy consumption. Therefore, further research on CCS disposal is required.

Glass-ceramics are a new type of inorganic material in which crystalline and glassy phases coexist, thereby combining typical glass and ceramic characteristics and improving the material performance and applicability. Solid waste can be used as a raw material to prepare high value-added glass-ceramics in which the heavy metal content in the waste is effectively solidified, which mitigates the environmental damage caused by HWIR and increases the economic value of solid waste. As such, solid wastes have attracted interest from researchers worldwide, with blast furnace slag, rice husk ash, coal fly ash, red mud and fly ash, electric arc furnace dust, and metallurgical silicon slag all explored as raw materials for producing glass-ceramics. Repurposing solid wastes for glass-ceramics reduces the overall volume of solid waste and enables the solidification of heavy metals. Zhang et al. proposed a new method involving the use of the metastable state of vitrified amorphous slag to solidify fly ash. By conducting long-
term leaching tests and assessing the potential ecological risk, the authors showed that the heavy metals in the sintered samples were stable and posed no direct threat to the environment. Alternatively, Chen et al. used melting method-induced sintering to prepare glass-ceramics based on SiO₂−Al₂O₃−Fe₂O₃−CaO incineration residue. This approach achieved high solidification efficiency (>99%) regarding the Zn, Cu, and Cr content of the incineration residue. However, Zhang et al. and Chen et al. both used solid wastes supplemented by various materials to prepare glass-ceramics, meaning that only a small amount of hazardous waste was used to solidify the heavy metals, thus limiting the amount of hazardous waste that was recycled and lowering the economic benefits of the process.

The preparation of glass-ceramics requires SiO₂, Al₂O₃, CaO, and Fe₂O₃, all of which are contained in HWIR. The principal components of CCS are summarized in Table 2, which lists relatively high concentrations of Pb (23.24%), Cr (12.90%), and Cu (10.95%). In addition, analytically pure acetic and nitric acids and deionized water were used in the leaching toxicity experiment.

2. MATERIALS AND METHODS

2.1. Materials. The HWIR and CCS used in the study were supplied by an environmental protection company in Chongqing, China. The main components of the HWIR were measured using X-ray fluorescence (XRF, see Table 1), with Si, Al, and Ca in the forms of SiO₂, CaO, and Al₂O₃, respectively, representing the most abundant elements. Then, the Cr, Fe, and Ni in CCS are a crystal nucleating agent that promotes the formation of crystal nuclei in glass-ceramics. Therefore, cosintering HWIR and CCS to realize solidification/stabilization of heavy metal contaminants reduces the accumulation of waste (e.g., in landfills) and environmental risk and can also be used to produce building materials.

By combining HWIR and CCS for treatment, this article focuses on recycling incineration residue to produce glass-ceramics and optimizing the heat treatment process through orthogonal experiments to maximize the quality of the recycled product for use as construction materials. We cosintered the CCS and an incineration residue base glass mixture to achieve a solidified product for use as construction materials. We cosintered the orthogonal experiments to maximize the quality of the recycled ceramics and optimizing the heat treatment process through focus on recycling incineration residue to produce glass-ceramics and their solidified bodies. In addition, the microstructures of the prepared glass-ceramics were analyzed using X-ray diffraction (XRD). Together, the microstructural and macroscopic properties of the produced glass-ceramics were used to elucidate the mechanism of heavy metal solidification in the HWIR and CCS.

2.2. Preparation of Incineration Residue-Based Glass-Ceramics. The incineration residue was dried, crushed or ball-milled, sieved (200 mesh), and then placed in a corundum crucible. The temperature was increased from room temperature to 1450 °C in a muffle furnace at a heating rate of 10 °C/min for 1 h to ensure complete melting of the sample. The corundum crucible was removed with crucible tongs, and the molten liquid was quickly poured into cold water to obtain the base glass. Next, the base glass was dried, ground, and sieved (200 mesh) to prepare it for use. Then, 3 g of the dried and sieved basic product was placed in a cylindrical corundum crucible mold, the diameter and height of which were both 20 mm. The mold was agitated gently to reduce the gaps between the powder particles and to ensure a flat sample surface. Then, the mold containing the sample was placed in a muffle furnace, heated to the nucleation temperature at a rate of 8 °C/min, maintained at this temperature for a certain period (orthogonal

### Table 1. Chemical Components of Hazardous Waste Incineration Residue (wt %)

|   | O   | Si  | Fe  | Al  | Ca  | Ti  | Na  | Zn  | P   | Mg  | K   | Other |
|---|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-------|
|   | 22.54 | 22.55 | 8.98 | 5.97 | 5.43 | 4.46 | 3.49 | 1.88 | 1.36 | 1.19 | 1.12 | 2.28   |

### Table 2. Chemical Components of Chromium-Containing Sludge (wt %)

|      | Pb | O   | Cr  | Fe  | Cu  | Na  | Cl  | S   | Ba  | Sn  | Ca  | Other |
|------|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|-------|
|      | 23.24 | 22.11 | 12.90 | 9.74 | 10.95 | 5.35 | 5.46 | 2.47 | 4.00 | 1.9  | 0.79  | 1.09   |

### Table 3. Orthogonal Experiment Design and Corresponding Compressive Strength Results

|   | no. | nucleation temperature (°C) | nucleation time (h) | crystallization temperature (°C) | crystallization time (h) | compressive strength (MPa) |
|---|-----|-----------------------------|---------------------|---------------------------------|--------------------------|---------------------------|
| 1 | 1   | 1 (740)                     | 1 (0.5)             | 1 (1060)                        | 1 (0.5)                  | 75.23                     |
| 2 | 1   | 2 (1080)                    | 2 (1)               | 2 (1479)                        | 2 (3)                    | 147.96                    |
| 3 | 1   | 1 (1100)                    | 1 (3.5)             | 3 (1181)                        | 3 (9.5)                  | 118.14                    |
| 4 | 2   | 2 (760)                     | 2                   | 3                               | 3                       | 95.11                     |
| 5 | 2   | 3 (780)                     | 3                   | 3                               | 3                       | 139.87                    |
| 6 | 2   | 3 (1.5)                     | 3                   | 2                               | 2                       | 258.73                    |
| 7 | 3   | 1                            | 3                   | 3                               | 2                       | 163.68                    |
| 8 | 3   | 2                            | 3                   | 1                               | 3                       | 141.25                    |
| 9 | 3   | 3                            | 3                   | 2                               | 1                       | 134.06                    |
| ki1|     | 113.78                      | 111.34              | 158.40                          | 116.39                   |                           |
| ki2|     | 164.57                      | 143.03              | 125.71                          | 190.12                   |                           |
| ki3|     | 146.33                      | 170.31              | 140.56                          | 118.17                   |                           |
| R  |     | 50.79                       | 58.97               | 32.69                           | 73.73                    |                           |
| optimal level | A2 | B3                           | C1                  | D2                              |                          |                           |

**Note:** $k_{ij}$ ($i = 1, 2, 3$) = 1/3 × summation of the test values at the same level; $R$, range value.
experiment), and then heated further to the crystallization temperature for a certain period (orthogonal experiment). The sintered product was cooled using the furnace to obtain the glass-ceramics. Based on previous experiments, four-factor, three-level orthogonal experiments were designed to explore the respective effects of nucleation temperature, nucleation time, crystallization temperature, and crystallization time on the resulting HWIR-based glass-ceramics for optimizing the preparation process. The design of the orthogonal experiment is presented in Table 3.

2.3. Preparation of Cosintered Solidified HWIR and CCS. A proportion of the incineration residue base glass was replaced by CCS, which had been dried, ball-milled, and sieved (200 mesh). Next, 3 g of the incineration residue base glass/CCS mixture was placed in a cylindrical corundum crucible mold (diameter, 20 mm; height, 20 mm), which was agitated gently to reduce the gaps between the powder particles and ensure surface smoothness. Then, the mold was placed in a muffle furnace, heated to the nucleation temperature at a rate of 8 °C/min, maintained at this temperature for a certain period, heated further to the crystallization temperature, and maintained at this new temperature for a certain period. The sintered product was cooled using the furnace to obtain a solidified product containing both HWIR and CCS.

2.4. Compressive Strength Measurements. The compressive strength of the samples was tested according to the GB/T17671-1999 standard. For each group, three parallel samples (relative error < 10%) were tested using a multifunction compressive strength testing instrument (AGN-250, Shimadzu), and the average compressive strength was taken as the final result.

2.5. Leaching Toxicity Tests. The TCLP was used to test the heavy metal leaching toxicity of the glass-ceramic samples relative to the US EPA standard. The pH of the extractant used in the TCLP was 2.88 ± 0.05. Prior to testing, 5.7 mL of glacial acetic acid was poured into a volumetric flask and diluted using deionized water to form a solution with a total volume of 1 L. The steps of the leaching toxicity tests are as follows: samples were crushed to produce particles with diameters < 9.5 mm; next, 0.5 g of the sample was added to an Erlenmeyer flask, followed by the addition of the extractant at a solid/liquid ratio of 1:10. After agitation, the Erlenmeyer flask was placed in a water bath horizontal oscillator (SHA-A, HuanYu Instrument) at constant temperature for shaking. The speed of the constant-temperature water bath horizontal oscillator was set to 120 r/min, the temperature was set to 25 ± 2 °C, and the oscillation time was set to 18 h. After shaking, the leaching solution was filtered, with the heavy metal leaching concentration of the sample determined using flame atomic absorption spectrometry (AA-6300C, Shimadzu). For each sample, three parallel samples were tested, and the average leaching concentration was taken as the final result. The total concentrations of heavy metals in the HWIR and CCS were analyzed by inductively coupled plasma-optical emission spectroscopy (ICP-OES) based on HJ-803-2016 (CN-HJ, 2016). The solidification efficiency was evaluated using the following formula

\[
\text{solidification efficiencies} = \frac{1}{\text{total mass of each toxic heavy metal in the initial solid}} \times \text{quality of each toxic heavy metal in the leaching solution}
\]

where the quality of each toxic heavy metal in the leaching solution is from the leaching test and the total mass of each toxic heavy metal is from the ICP-OES test based on HJ-803-2016.

2.6. Characterization Analyses. The thermal behavior of the incineration residue base glass was analyzed using differential scanning calorimetry (DSC; STA449F3, Netzsch). Testing was performed by increasing the temperature from room temperature to 1200 °C at a heating rate of 20 °C/min in air. The crystalline composition of the samples was determined by XRD (PANalytical B.V., The Netherlands) using Cu Kα radiation at 40 kV and 30 mA.

3. RESULTS AND DISCUSSION

3.1. DSC Curve of the HWIR Base Glass. Figure 1 shows that the HWIR base glass loses no weight below 1200 °C, with the glass transition temperature (\(T_g\)) appearing at 672 °C and the exothermic peak of crystallization spanning 870–1170 °C with the peak temperature (\(T_p\)) at 1080 °C. As the span of the exothermic peak is large (300 °C) and does not change dramatically, the powder sintering method is suitable for preparing glass-ceramics.

3.2. Heat-Treatment System Optimization for Incineration Residue-Based Glass-Ceramics. We have previously reported that under a crystallization temperature of 1080 °C, crystallization time of 1 h, nucleation temperature of 760 °C, and nucleation time of 1 h, the compressive strength of self-sintered glass-ceramics can reach 204.84 MPa. Compared with previous experiments, the gradient of the heat treatment system was reduced, indicating that the compressive strength can be used as an indicator when optimizing the heat treatment system (in terms of nucleation temperature, nucleation time, crystallization temperature, and crystallization time) through an orthogonal experiment. The experiment design and optimization results are presented in Table 3 and Figure 2, respectively. For each group, three parallel samples were tested, and the average compressive strength was taken as the final result. The error bar is the standard deviation of the three parallel samples.

As shown in Figure 2, the compressive strength of HWIR glass-ceramics first increases and then decreases. The nucleation temperature increases from 740 to 780 °C, reaching a maximum of 164.57 MPa at 760 °C. The nucleation
temperature is between 50 and 100 °C higher than the glass transition temperature because the viscosity in the glassy state is more conducive to the formation of a large number of nuclei at this temperature, providing a suitable prerequisite for subsequent crystal growth. When the crystallization temperature is within the temperature range of the endothermic peak, the compressive strength reaches a maximum value of 158.40 MPa at 1060 °C. As the temperature increases, the compressive strength of glass-ceramics decreases because the crystal grains grow abnormally when the crystallization temperature is too high. However, increasing the temperature further causes the grains to melt, which is not conducive to a uniform and dense grain distribution and results in decreasing the density and compressive strength.

Regarding the nucleation and crystallization times, the compressive strength peaked at 1.5 and 1.0 h, respectively. Some studies have shown that the degree of crystallinity determines mechanical strength. Increasing the nucleation time causes the number of crystal nuclei to increase continuously under an appropriate glass viscosity, thus providing the basis for subsequent crystal growth. In addition, the compressive strength first increases and then decreases as the crystallization time increases from 0.5 to 1.5 h, reaching its highest value at 1 h. This may be because crystal growth is not completed when the crystallization time is short. For complete crystal growth after 1 h, the compressive strength increases; however, for polycrystalline materials, signs of damage, including fractures, appear at the grain boundaries, which indicates that smaller grain size and tighter arrangement correspond to greater mechanical strength. As the crystallization time increases further, the grains become gradually larger, thereby decreasing the compressive strength.

The range analysis of the orthogonal experiment elucidates the order of influence (from greatest to least) of the four factors affecting the compressive strength of HWIR-based glass-ceramics: crystallization time > nucleation time > nucleation temperature > crystallization temperature. In addition, A2, B3, C1, and D2 were identified as the superior levels of A, B, C, and D, that is, a nucleation temperature of 760 °C, nucleation time of 1.5 h, crystallization temperature of 1060 °C, and crystallization time of 1 h. Under these conditions, the compressive strength of the HWIR-based glass-ceramics reached 258.73 MPa.

3.3. Cosintering of HWIR and CCS to Solidify Heavy Metals. Owing to the activity of the HWIR base glass, we decided to replace part of it (10, 20, 30, 40, and 50%) with CCS to prepare glass-ceramics and realize CCS solidification. For each group, three parallel samples were tested, and the average compressive strength was taken as the final result. The error bar is the standard deviation of the three parallel samples.

![Figure 2. Influence of various factors on the compressive strength of glass-ceramics: (A) nucleation temperature, (B) nucleation time, (C) crystallization temperature, and (D) crystallization time.](https://doi.org/10.1021/acsomega.1c01659)
The influence of the CCS content percentage on the compressive strength of the solidified body is shown in Figure 3, which shows that the compressive strength of the combined HWIR-CCS solidified body first increases and then decreases with an increase in the CCS content. Compared with the purely HWIR-based glass-ceramics, the addition of CCS noticeably reduces the compressive strength; for example, the compressive strength of the IRCS sample containing 10% CCS is 115.79 MPa. This can be attributed to the dominance of the crystalline phase and low Ca and Si content in CCS, with the latter being the key element for preparing glass-ceramics. In response to increasing the CCS content, the compressive strength of IRCS samples gradually increases, reaching a maximum of 142.68 MPa for a CCS content of 30%. This result can be explained by the presence of Cr₂O₃ and Fe₂O₃ nucleators of glass-ceramics, in the CCS, which promotes crystal growth and enhances the compressive strength of the solidified body. However, CCS is mainly in a low-activity crystalline phase, which can be seen in the XRD section. In addition, the curing capacity of HWIR-based glass-ceramics is limited. Reducing the HWIR base glass weakens the curing effect, leading to the compressive strength decreasing at the macroscopic scale. Nevertheless, the IRCS sample containing 50% CCS still registered a compressive strength of 89.56 MPa, which comfortably meets the requirement for construction materials (>0.3 MPa). The influence of the CCS content percentage on the compressive strength of combined HWIR-CCS solidified bodies with increasing CCS content.

HWIR-CCS (IRCS) solidified body first increases and then decreases with an increase in the CCS content. Compared with the purely HWIR-based glass-ceramics, the addition of CCS noticeably reduces the compressive strength; for example, the compressive strength of the IRCS sample containing 10% CCS is 115.79 MPa. This can be attributed to the dominance of the crystalline phase and low Ca and Si content in CCS, with the latter being the key element for preparing glass-ceramics. In response to increasing the CCS content, the compressive strength of IRCS samples gradually increases, reaching a maximum of 142.68 MPa for a CCS content of 30%. This result can be explained by the presence of Cr₂O₃ and Fe₂O₃ nucleators of glass-ceramics, in the CCS, which promotes crystal growth and enhances the compressive strength of the solidified body. However, CCS is mainly in a low-activity crystalline phase, which can be seen in the XRD section. In addition, the curing capacity of HWIR-based glass-ceramics is limited. Reducing the HWIR base glass weakens the curing effect, leading to the compressive strength decreasing at the macroscopic scale. Nevertheless, the IRCS sample containing 50% CCS still registered a compressive strength of 89.56 MPa, which comfortably meets the requirement for construction materials (>0.3 MPa).

Unsurprisingly, given the presence of Cr, Pb, and Cu in CCS, the leaching concentrations of Cr, Pb, and Cu increased with increasing CCS content. In addition, the corresponding decrease in HWIR caused the curing ability of the glass-ceramics to decline. Regarding Cu and Cr, their leaching concentrations were less than the US EPA limits (Cu: 15 mg/L, Cr: 5 mg/L) when the CCS contents were 20 and 30%, respectively. However, when the CCS content was 10%, the Pb leaching concentration was 9.96 mg/L, which exceeds the US EPA standard of 5 mg/L. The Pb leaching concentration is known to increase in response to a decrease in the Si, Mn, and Fe contents. Therefore, as increasing the CCS content reduces the amount of Si, the increase in Pb leaching is not surprising.

Figure 4 shows the solidification efficiency of the various heavy metals in the IRCS samples. The solidification efficiencies of Cr, Cu, and Pb were all greater than 90%, with the Cr and Cu efficiencies exceeding 99%. Increasing the CCS content caused the solidification efficiencies of Cr and Cu to increase and then to decrease, exhibiting the same trend as observed for the compressive strength. Increasing the compressive strength restricted heavy metal leaching, thereby increasing the curing efficiency of Cr and Cu in the solidified body. Rigorous leaching concentration and solidification efficiency analyses showed that glass-ceramic samples prepared by cosintering under specific heat treatment conditions yield desirable results regarding the solidification of heavy metals, such as Cr, Pb, and Cu.

While Cr exists predominantly in forms of Cr(III) and Cr(VI), the latter presents the greatest toxicity risk to humans. The Cr(VI) leaching concentrations for the solidified bodies with different CCS contents are shown in Figure 6. Compared with the initial CCS leaching concentration, the Cr(VI) leaching concentration of the IRCS samples first decreased and then gradually increased as the CCS content increased, with minimum Cr(VI) leaching coinciding with a CCS content of 10%.

Table 4. Leaching Concentrations of HWIR and CCS

| heavy metal | Cu (mg/L) | Zn (mg/L) | Cr (mg/L) | Pb (mg/L) |
|-------------|-----------|-----------|-----------|-----------|
| HWIR        | 15.96     | 70.60     | 0.98      | 1.14      |
| CCS         | 6960.75   | 4840.88   | 4.8       |           |
| US EPA limit| 15        | 5         | 5         |           |

Although the US EPA has no relevant standard limit for Zn, the GBS085.3-2007 critical limits (China) state a Zn limit of 100 mg/L (>70.60 mg/L). Therefore, the subsequent leaching concentrations consider the heavy metals Cr, Cu, and Pb only. With reference to the CCS content, the tested samples were denoted as IRCS-0 (0%), IRCS-10 (10%), IRCS-20 (20%), IRCS-30 (30%), IRCS-40 (40%), and IRCS-50 (50%). The leaching results are presented in Figure 4. It should be noted that both the leaching concentration and curing efficiency reflect the curing ability; therefore, we considered the cosintering solidification process of HWIR and CCS in terms of both leaching concentration and solidification efficiency.

Compared with the initial leaching results of the HWIR and CCS, the heavy metal leaching concentration was significantly reduced for the cosintered and solidified IRCS samples. Unsurprisingly, given the presence of Cr, Pb, and Cu in CCS, the leaching concentrations of Cr, Pb, and Cu increased with increasing CCS content. In addition, the corresponding decrease in HWIR caused the curing ability of the glass-ceramics to decline. Regarding Cu and Cr, their leaching concentrations were less than the US EPA limits (Cu: 15 mg/L, Cr: 5 mg/L) when the CCS contents were 20 and 30%, respectively. However, when the CCS content was 10%, the Pb leaching concentration was 9.96 mg/L, which exceeds the US EPA standard of 5 mg/L. The Pb leaching concentration is known to increase in response to a decrease in the Si, Mn, and Fe contents. Therefore, as increasing the CCS content reduces the amount of Si, the increase in Pb leaching is not surprising.
pure CCS sample, the leaching concentration of Cr for each IRCS sample was significantly lower and, most importantly, less than the relevant limit (5 mg/L) associated with the identification standard for hazardous waste leaching toxicity identification (GB5085.3-2007, China), and less than the technical specification for environmental protection from chromium residue pollution control (HJ/T 301-2007, China). For Cr residues in industrial solid waste for landfill, the Cr(VI) limit was 3 mg/L, with our results showing that Cr(VI) leaching from IRCS solidified bodies with a CCS content below 50% meets this requirement. In conclusion, HWIR-based glass-ceramics are effective at solidifying the Cr(VI) content in CCS. According to the toxicity results regarding Cr(VI) leaching, although the total chromium leaching concentration for the IRCS-30 sample exceeded the relevant standard of 5 mg/L, the leached product consisted primarily of Cr(III), which is far less toxic.

3.5. X-ray Diffraction Analysis of Heavy Metal Solidification. The XRD patterns of HWIR, CCS, IRCS-0, IRCS-10, IRCS-20, IRCS-30, IRCS-40, and IRCS-50 were analyzed using MDI JADE software (version 6.5, see Figures 7 and 8). Figure 7 shows a broad weak peak spanning approximately 15°–40°, indicating that HWIR is an amorphous material. However, the XRD pattern of CSS corresponds to that of crocoite (PbCrO₄), which does not contain active minerals, such as glass.

The XRD patterns of the IRCS samples are shown in Figure 8, indicating that the main crystals precipitated from the self-sintering of HWIR glass-ceramics are hematite (Fe₂O₃, PDF #89-2810), ferrobrookite (Fe₂TiO₅, PDF #76-1743), and anorthite (Ca(Al₂Si₂O₈), PDF #89-1462). In particular, hematite and ferrobrookite have more diffraction peaks that span a wider angle range than anorthite. The precipitation and growth of the crystals within the sample determine its macroscopic properties, with the good crystal structure of...
the glass-ceramics confirming the excellent physical properties of the glass-ceramics at the microscopic level.\(^{21,32}\) Increasing the CCS content leads to a gradual decline in the crystal phase strength of hematite because of the change in sample composition. At an IRCS–CSS mixing ratio of 20%, the increased Cr and Pb proportions induce new chromite (\(\text{FeCr}_2\text{O}_4\), PDF #34-0140) and magnetite (\(\text{PbFe}_2\text{O}_4\), PDF #75-1632) crystal phases owing to the partial transformation of hematite. Increasing the CCS content further increases the intensity of the diffraction peaks corresponding to galena and chromite. When the CCS content reaches 50%, anorthite (\((\text{Ca}_4\text{Al}_2\text{Si}_6\text{O}_{16})\text{Cl}_2\), PDF #89-1462) transforms into sodalite ((\((\text{Na}_4\text{Ca}_4\text{Al}_6\text{Si}_{18}\text{O}_{48})\text{Cl}_2\), PDF #82-1811) as a consequence of an increase in Cl contributed by the CCS. Comparing the crystalline phase composition of the IRCS solidified bodies with different CCS contents reveals that certain heavy metals drive the formation of the crystal lattice (\(\text{FeCr}_2\text{O}_4\), \(\text{PbFe}_2\text{O}_4\)), explaining the decrease in their leaching concentrations. In addition, the emergence of the heavy-metal-based crystal phase at a CCS content of 20% coincides with the peak Cr and Pb curing efficiencies. As the CCS increases, the heavy metal content increases accordingly. Although certain heavy metals participate more notably in crystal formation, because their content is limited, the overall leaching concentration increases; however, the solidification efficiency remains high.

4. CONCLUSIONS

In this study, we used HWIR as the raw material to obtain base glass via a combination of high-temperature melting and water quenching. Subsequently, the HWIR-based glass-ceramics were prepared via sintering and solidification. Owing to the strong sintering activity and crystallization ability of the HWIR base glass, we proposed a method of cosintering and solidifying the heavy metals in HWIR and CCS to facilitate recycling of HWIR and CCS while mitigating the risk of secondary pollution.

The physicochemical compositions of HWIR and CCS were analyzed by XRF and XRD. The results showed that the HWIR was rich in SiO\(_2\), Fe\(_2\)O\(_3\), Al\(_2\)O\(_3\), CaO, and TiO\(_2\), providing the basis for preparation of glass-ceramics. Four factors, namely, crystallization temperature, crystallization time, nucleation temperature, and nucleation time, were selected for analysis via an orthogonal experiment. We determined that the optimal heat treatment system comprises a nucleation temperature, nucleation time, crystallization temperature, and crystallization time of 760 °C, 1.5 h, 1060 °C, and 1 h, respectively. Under these conditions, the compressive strength of glass-ceramics can reach 258.73 MPa.

We used HWIR as the raw material to solidify CCS. Six HWIR-CCS mixing ratios (0, 10, 20, 30, 40, and 50%) were selected for the solidification experiments. The experimental results show that the compressive strength of the solidified CCS first increases and then decreases as the CCS content increases. Moreover, the leaching toxicity of heavy metals (Cr, Pb, and Cu) increases as the CCS content increases. The Cu and Cr leaching concentrations exceed the standard limits when the CCS content reaches 20 and 30%, respectively. However, owing to the effect of the Si, Mn, and Fe content on Pb leaching, it behaves differently from the leaching of Cr and Cu. Based on the total amount of heavy metals in the samples and heavy metal leaching concentrations, the curing efficiencies of Cu, Cr, and Pb all exceeded 90%. Combined with the formation of heavy metal crystalline phases, as revealed by XRD analysis, certain heavy metals participate in the formation of the crystal lattice (\(\text{FeCr}_2\text{O}_4\), \(\text{PbFe}_2\text{O}_4\)), thus solidifying the heavy metals in the crystal lattice and reducing the overall leaching concentration. Therefore, these results demonstrate that the cosintering of HWIR and CCS can realize heavy metal solidification effectively. This study highlights the potential of the proposed method as a safe approach for removing HWIR and CCS, which could have a significant impact on reducing the environmental pollution caused by industrial processes.

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