

Article

Study on the Long-Term Durability and Leaching Characteristics of Low-Consumption Cement Backfill under Different Environmental Conditions

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Abstract: The high consumption and high cost of cement are the bottleneck problems that limit the development of cemented tailings backfilling technology. The low-consumption cement backfill is immersed in a weak acid/alkaline groundwater environment for a long time. Reducing the consumption of cement can easily lead to problems such as a sudden decrease in strength and the leakage of heavy metals. Through the monolithic leaching test in static and uniaxial compressive tests, the heavy metals' leaching concentration and the compressive strength of low-consumption cement backfills in different pH soaking solutions were measured at different soaking times. Results show that a lower cement concentration will result in a lower CTB compressive strength and a higher rate of heavy metal leaching. Long-term exposure to an acidic/alkaline environment will lead to the instability and destruction of the CTB structure. A microscopic examination reveals that the creation of hydration products can improve the structure's compactness while also lowering the internal porosity of CTB but can also solidify heavy metal ions in various ways. A first-order reaction/diffusion model (FRDM) can better evaluate the leaching behavior of CTB. This study helps to improve backfilling technology, thereby contributing to the creation of sustainable mining geotechnologies.

Keywords: cementing backfilling technology; cement consumption; heavy metal leaching; pollution prevention; structural safety; balanced development



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1. Introduction

With the promotion of green and sustainable development in global mines as a novel technology to achieve environmentally friendly and sustainable mining resource development, cemented tailings backfilling technology has been continuously improved and promoted and has become the preferred way to deal with tailings [1–3]. However, long-term immersion of cemented tailings backfill (CTB) in groundwater environments can easily lead to heavy metal leakage and structural damage, posing potential hazards to the ecological environment and mine safety. Solving this problem has become a major challenge for current backfilling technology [4–6].

The tailings' surface contains active heavy metal ions. In addition to being used for particle refinement, the mechanical energy applied by crushing is also stored in the particle

system, leading to flaws, amorphousness, free radical production in the particle lattice, and a surface free-energy increase, which promotes the activity of heavy metal elements and enhances the reaction force [7–9]. The process of backfilling underground carries a danger of environmental contamination [10]. As the main raw material of CTB, cement cannot only play the role of cementation, but it can also effectively solidify/stabilize toxic substances [11,12]. When cement consumption decreases, the strength of the backfilling decreases, and heavy metal pollution raises the danger of environmental safety when the ability of heavy metal ions to solidify declines, causing the surrounding rock to become unstable [13,14]. The primary factor influencing CTB strength and heavy metal ions' solidification is cement usage [15]. Suwimol Asavapisit et al. assessed the samples' unconfined compressive strength and examined the solidification processes of the fixed zinc cyanide plating sludge with varying concentrations of ordinary Portland cement (OPC) used as solidification binders. The findings demonstrate that as the cement content drops, the sample's strength drops dramatically, and that there are considerable variations in the heavy metals' leaching concentrations of Cr, Zn, and Fe [16]. Choon Keun Park used three different cementitious materials to investigate the capacity of hazardous wastes containing heavy metals to solidify or stabilize and evaluated the sample's compressive strength. The findings demonstrate that regular Portland cement (OPC) may successfully lower Cr, Cd, and Pb's leaching concentration and exhibits solidification/stabilization performance when used as a cementitious material sample [17]. M. Tyrer et al. discussed how cement and heavy metals interact throughout the solidification/stabilization process and concluded that cement is currently the most suitable adhesive for solidifying heavy metals [18]. The study by Rachana Malviya et al. demonstrates that heavy metals (Pb, Zn, Fe, Cu, and Mn) in hazardous sludge from steel production plants may be efficiently solidified by cement, and that the sample's unconfined compressive strength diminishes as the cement level does [19]. There is a large potential for a decrease in the dosage of cementitious materials. However, the decrease in cement consumption will inevitably lead to a weakening of the ability of the backfilling material to solidify/stabilize heavy metal ions [20]. Therefore, from the structural–environmental perspective, the “minimum strength of the CTB” and “effective control of heavy metal leaching and diffusion” have become the dual control standards for determining cement consumption under safe conditions.

The structural impact on surrounding rock control and the environmental impact on heavy metal ion solidification may be used to sum up the effects of CTB [21,22]. On the one hand, CTB is in weak acid/alkali mine water for a long time, and the damage of acid/alkali erosion to the CTB structure and the leaching of heavy metal ions are weak, which is one of the main theoretical bottlenecks restricting the reduction of cementitious materials. Joris J. Dijkstra. investigated the impact of leaching solution with a pH value in the range of 0.4–12 on the static leaching test's effects on heavy metals' leaching (Ni, Cu, Zn, Cd, and Pb) in polluted soil. There was a discovery of a significant pH dependency in heavy metals' leaching [23]. Souhail R. Al Abed et al. conducted research on how pH affected heavy metal discharge from mineral wastes. Cu, Pb, and Zn leach more quickly in acidic environments, but in alkaline environments, Cu, Pb, and Zn precipitate as pH rises. The impact of heavy metal leaching varies depending on the pH level [24]. Andreas Voegelin et al. looked at the release of Zn, Cd, Pb, and Cu from four polluted soils using single- and multiple-batch extractions together with column leaching. They discovered that in two soils with pH values that were almost neutral, the release amounts of zinc and copper were significantly lower. However, when pH was adjusted to 3, they found that concentrations of the four heavy metals in the polluted soil, Zn, Cd, Pb, and Cu, were decreased by 65% to 90% [25]. On the other hand, reducing the content of cementitious materials led to a decrease in nano-scale pores in CTB, a decrease in the compactness of hydration products, an intensification of the seepage process and an expansion of big pores, a decrease in adsorption performance, and a decrease in the lattice capacity of the chemical reaction due to the decrease in the number of hydration products [26,27]. The solidification ability of CTB toward heavy metal ions is changed from traditional “strong solidification”

to “weak solidification”, and the research on the solidification ability of the low strength of CTB has not been carried out [28,29].

Therefore, this paper designs CTB using different cement-to-sand ratios to reduce cement consumption and carry out a static leaching test to reveal the strength change of low-consumption cement backfill and the properties of four heavy metals that leach from the perspective of chemistry and mechanics. Simultaneously, heavy metals’ physical and chemical solidification mechanism by different types of hydration products and microstructures were analyzed by means of SEM scanning, XRD detection, and other technical means, so as to explore the mechanical behavior and leaching characteristics of different degrees of acid/alkali erosion on low-strength CTB. Finally, to assess the leaching kinetics of heavy metal components, a first-order reaction/diffusion model was developed. This study not only pays attention to the strength change of CTB under the influence of cement consumption, but it also focuses on the environmental risk of metal leaching in CTB. This not only helps to prevent and control heavy metal pollution in tailings backfilling, but it also provides a development idea for reducing the actual demand of cement consumption in backfilling technology. At the same time, it is helpful to break through the bottleneck of the balanced development concept of the “safety-environment-economic” effect of backfilling technology and encourage green mines’ sustainable development.

2. Experiment

2.1. Material

The main materials of CTB are cement, tailings, and water.

2.1.1. Cement

Ordinary Portland cement of P.O. 42.5 used in this experiment was produced by Jiaozuo Qianye New Material Co., Ltd. (Jiaozuo, China). As indicated by Tables 1 and 2, the primary physical performance metrics and chemical composition were examined. The performance metrics satisfy the “General Portland Cement” national standard’s standards (GB175-2007).

Table 1. Main chemical components.

Mineral Component	CaO	SiO ₂	Al ₂ O ₃	Fe ₂ O	Na ₂ O	K ₂ O
Mass percentage (%)	65	22	6.5	3.2	0.18	0.34

Table 2. Physical performance indicators.

Specific Surface Area/(m ² /kg)	Ignition Loss	Stability	Setting Time/Min		Compressive Strength/MPa		Bending Strength/MPa	
			Initial Set	Final Set	3 d	28 d	3 d	28 d
379	4.00	No cracks and no warping were found.	176	236	28.6	43.2	5.7	8.5

2.1.2. Tailings and Water

The entire set of tailings comes from a Shandong Province gold mine. The physical characteristics of tailings include specific surface area, bulk density, porosity, and water content, which are shown in Table 3. Figure 1 shows that the particle size distribution is measured. Tailings particles belong to medium-coarse unclassified tailings, of which particles above 0.074 mm account for 57.13%, the median particle size is 0.0798 mm, and the tailings particle size is moderate. Using XRD technique, the gold tailings’ phase analysis was completed (see Figure 2). The gold tailings mainly contain several main mineral components such as muscovite, quartz, and albite. In addition, gypsum, pyrite, and other mineral phases were also detected. Table 4 displays semi-quantitative content. The appearance of tailings particle surfaces was observed by SEM at 3200× and 5500×

magnification in Figure 3. The majority of the particles' surface is made up of some lamellar and irregular layered structures which were rough and porous, with many tiny voids. The large particles are mostly micron-sized fine particles, with irregular and uneven shapes and sharp edges. Gold tailings' primary chemical makeup, which is mostly made up of Si and Al components with trace quantities of other metal elements, was identified using XRF. These elements are stored in different minerals in the form of oxides, and the corresponding components are shown in Table 5.

Table 3. Physical properties of gold tailings.

Material	Moisture Content/%	Relative Density/(g/cm ³)	Bulk Density/(g/cm ³)	Bulk Density/(g/cm ³)	Specific Surface Area/(m ² /g)
Gold tailings	25.82	2.56	1.52	40.6	4869

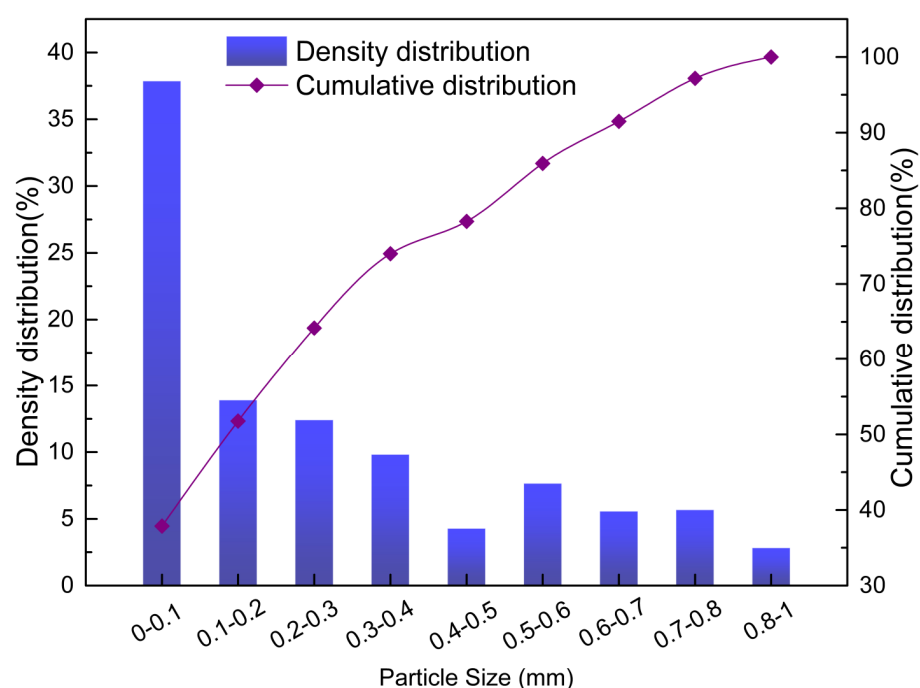


Figure 1. Tailings particle size distribution map shapes and sharp edges.

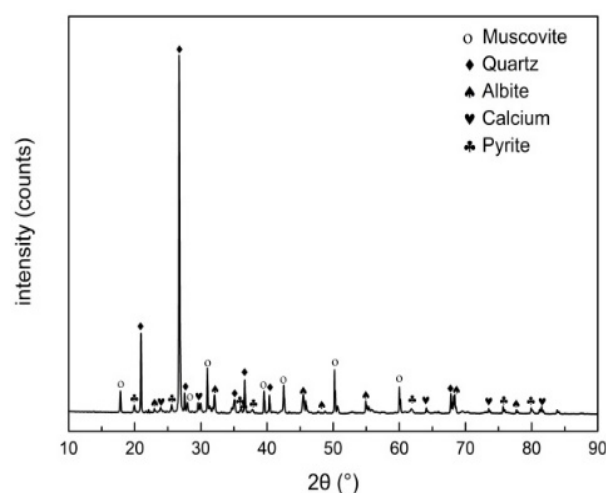
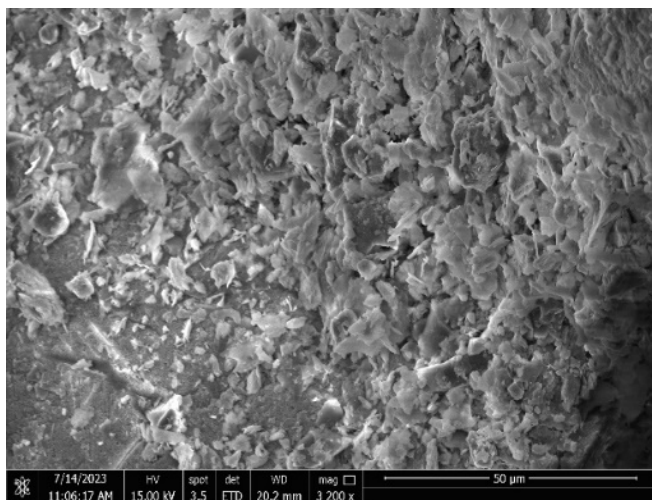
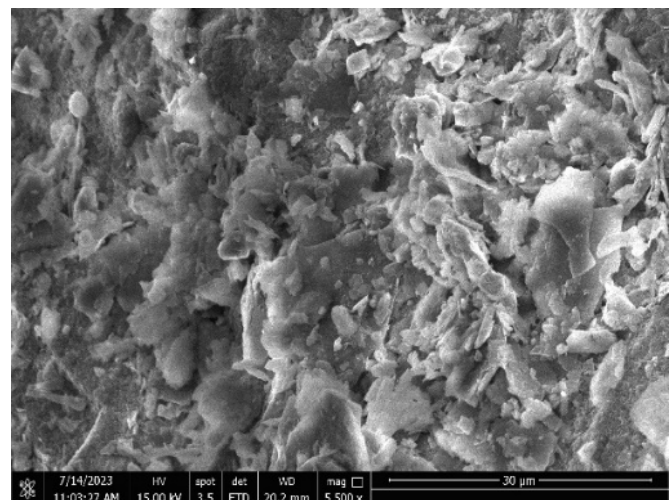


Figure 2. XRD of gold tailings.

Table 4. Mineral composition of gold tailings.

Mineral	Chemical Formula	Semi-Quantitative
Muscovite	$\text{CaMg}(\text{CO}_3)_2$	42%
Quartz	SiO_2	35%
Albite	$\text{Na}[\text{AlSi}_3\text{O}_8]\text{-Ca}[\text{Al}_2\text{Si}_2\text{O}_8]$	14%
Gypsum	CaCO_3	7%
Pyrite	FeS_2	2%

**(a)** 3200× magnification**(b)** 5500× magnification**Figure 3.** SEM images of gold tailing of different magnification: (a) 3200× magnification; (b) 5500× magnification.**Table 5.** The main chemical composition of gold tailings.

Chemical Composition	SiO_2	Al_2O_3	K_2O	Fe_2O_3	LOI	MgO	CaO	TiO_2	MnO	P_2O_5	Na_2O
Content/%	80.96	11.09	3.82	1.94	1.3	0.37	0.31	0.12	0.03	0.04	0.34

Ordinary tap water was used to make the specimen, and the deionized water was selected for the heavy metal leaching test.

2.2. Sample Preparation

According to the actual requirements of tailings backfilling, three groups of slurry concentrations with low cement consumption were selected with the mass fraction of 72% and the ratio of 1:10, 1:15, and 1:20 for cement–sand, respectively.

The required mass of cement, tailings, and water was weighed by electronic weighing and added to the mixer for full mixing. Then, the backfilling slurry was injected into the 70.7 mm × 70.7 mm × 70.7 mm standard triple test mold. After compaction, vibration, and smoothing. It was placed in humid air with constant temperature ($20 \pm 5^\circ$) and constant humidity (more than 90%) for maintenance. After the slurry was formed, the demoulding was carried out, and the group number and date were all numbered. The process is shown in Figure 4.

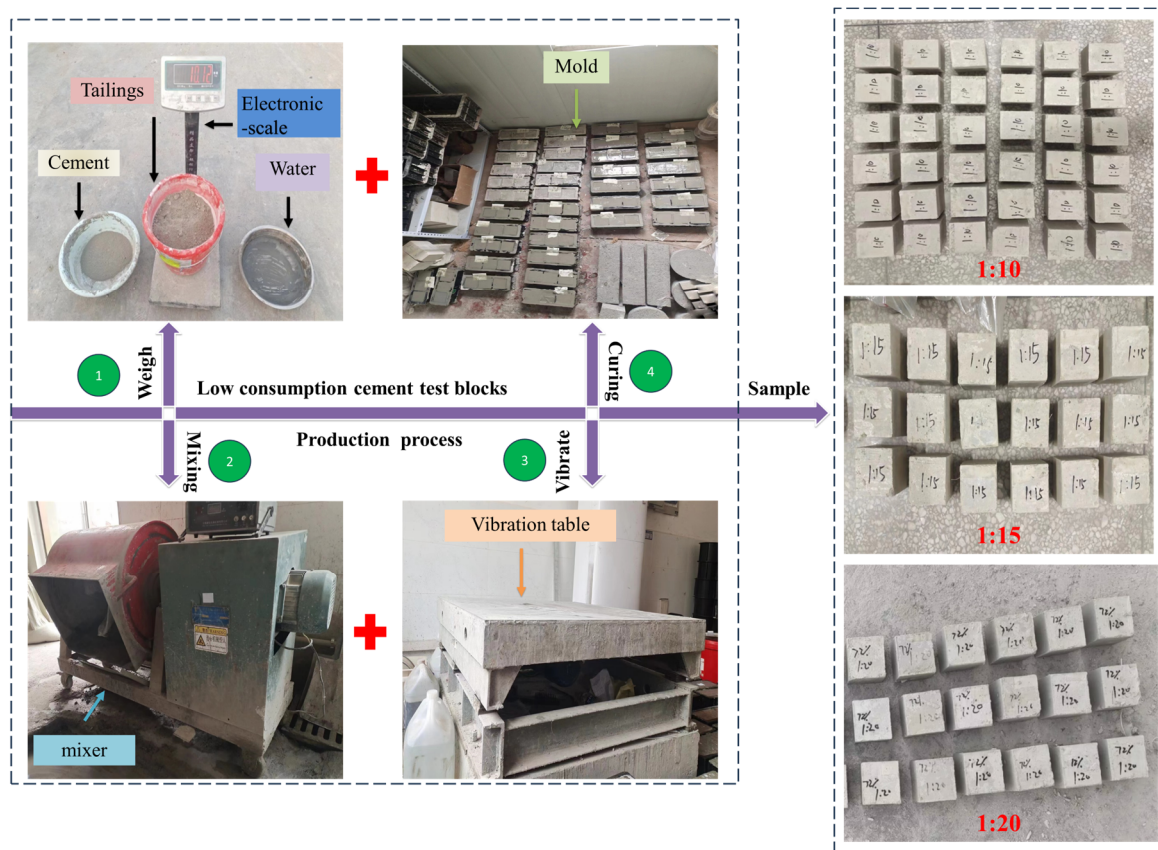


Figure 4. Cube specimen production process.

2.3. Testing Method

2.3.1. ICP-AES Test

ICP-AES test refers to the solid waste determination of 22 metal elements—inductively coupled plasma–optical emission spectrometry test method. To measure the content of heavy metals in the sample, it is necessary to establish the corresponding standard curves of four heavy metal elements. The mixed standard solutions of heavy metals with concentrations of 1 ppm, 2 ppm, 3 ppm, 4 ppm, and 5 ppm were prepared and introduced into ICP-AES in turn to measure the emission intensity to obtain the concentration–intensity standard curve of the corresponding heavy metals. The leaching solution of the specimens collected at 10 d, 20 d, 30 d, 60 d, 90 d, and 120 d was introduced into ICP-AES to measure the content of heavy metals Cu, Zn, Pb, and Ni.

2.3.2. Static Leaching Test

Refer to the United States Environmental Protection Agency (EPA) SW-846 1315 test method, Leaching Method of Solid Waste—Sulfuric Acid and Nitric Acid method (HJ/T299-2007), German standard leaching method DIN 38414-S4, and other solid leaching test standards, according to the actual situation, and adjust the leaching test conditions appropriately. Simulated downhole acid and weak alkali environment and soaking solutions with pH of 4, 6, 7, 8, and 10 were prepared by stepwise dilution method. The leaching test container adopts a round-shaped polypropylene plastic transparent round box, so that the six sides of the test cube can fully have contact with solutions. Soaking solutions submerge the specimen solidified body at a height of not less than 20 mm, and the corresponding information label is marked on the periphery of the container.

The three kinds of cement–sand ratio test cube (1:10, 1:15, 1:20) were placed in the soaking solution with different pH values (4, 6, 7, 8, 10), and an appropriate amount of solution samples were taken at the specified soaking time (10 d, 20 d, 30 d, 60 d, 90 d, 120 d)

and then filtered with the 0.45 μm membrane filter and stored in a centrifuge tube with a volume of 10 mL. In total, 1% nitric acid was added to the filtrate, and leaching values of four heavy metal ions were obtained by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES). During the experiment, the pH value was monitored twice a day, and nitric acid that had been diluted was added to the solution to modify the pH value and maintain it at a set value.

2.3.3. Uniaxial Compression Test

A straightforward and widely used technique for measuring the compressive strength of CTB is the uniaxial compressive strength test (UCS). UCS test used a hydraulic universal testing machine (WEW-600B, CHN, Jinan, China), and the displacement loading method was used to test the strength. When the soaking time of the specimen reached 30 d, 60 d, 90 d, and 120 d, respectively, Cubes' compressive strength was tested by displacement load method, repeated three times, and the average value was calculated.

2.3.4. SEM Test

The microstructure of the samples at different hydration times was observed by scanning electron microscope (Quanta 250 FEG, Washington, DC, USA). The center of the specimen after crushing in the uniaxial compression test was sampled. And using a tool knife, a little piece of approximately 3 mm in diameter and 1 mm in thickness was formed. The test surface was polished and leveled, dried and sprayed with gold, and then vacuum-tested on the SEM instrument.

2.3.5. XRD Test

The mineral composition of hydration products was analyzed by XRD (Smart Lab, JPN, Hong Kong). After the compressive strength test, the center of the specimen was sampled and the hydration of the model was stopped with anhydrous ethanol. The test angle is scanned between 10° and 90° . Both step size and emissivity are constant.

3. Result and Discussion

3.1. The Uniaxial Compressive Strength of CTB

Figure 5 illustrates how the compressive strength of CTB changed. In general, with the increase in curing time, the compressive strength of the CTB test cube with different cement–sand ratios in the same pH solution is generally the same, but compressive strength decreases as the cement–sand ratio increases. This is because, in the case of the same slurry mass fraction, the larger the cement–sand ratio is, the more cement is used; the higher the hydration reaction efficiency is, the greater the production of hydration products; and the strength of CTB is improved within a certain range [30].

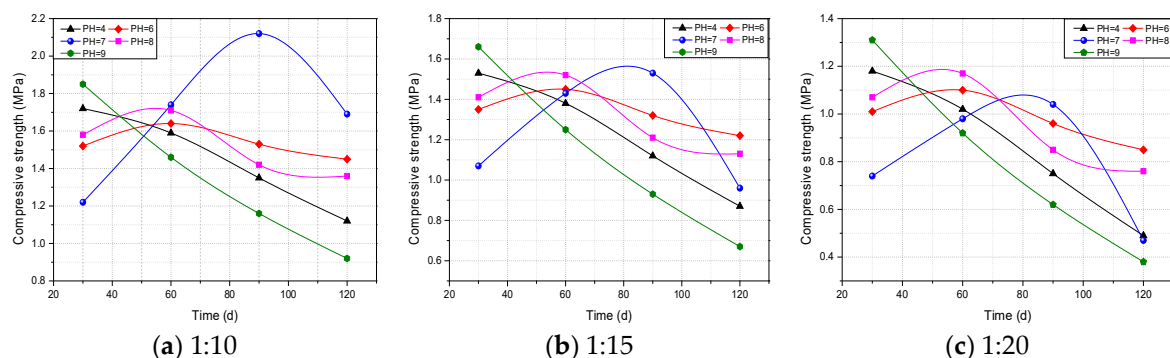


Figure 5. Compressive strength of CTB with different cement–sand ratios are soaked for different times. (a) Compressive strength of CTB with cement–sand ratio of 1:10 soaked for 30, 60, 90, 120 days. (b) Compressive strength of CTB with cement–sand ratio of 1:15 soaked for 30, 60, 90, 120 days. (c) Compressive strength of CTB with cement–sand ratio of 1:20 soaked for 30, 60, 90, 120 days.

The compressive strength of CTB with three different cement–sand ratios has a common change rule in the same acid/alkaline environment. In the solution of pH = 4, the CTB's compressive strength reached the maximum value within 30 days, and then the compressive strength decreased almost linearly. In the solution of pH = 6, the compressive strength of CTB continued to increase after 30 days, and it reached its maximum within 60 days. After that, the strength began to decrease gradually. In an alkaline solution, the compressive strength change in CTB is similar to that in an acidic solution. The strength change increases first and then decreases with the increase in soaking time, and the stronger the alkalinity, the shorter the time for the compressive strength to reach the maximum value, and thereafter, the more the strength declines. Within 30 to 90 days, the UCS increased further in pH = 7, and it continued to decline from 90 d to 120 d. In the long term, the UCS of CTB in a neutral environment is greater than that in an acidic environment and alkaline environment. In the short term, CTB's compressive strength in an alkaline environment is slightly larger than that in an acidic environment. This is mainly because CTB will accelerate the internal hydration reaction in an acidic/alkaline environment, generate more hydrated calcium silicate (C-S-H), and improve the early cementation strength [31,32].

Meanwhile, SO_4^{2-} in acidic solution and $\text{Ca}(\text{OH})_2$ products in CTB continuously fill the internal pores of CTB, while OH^- in the alkaline environment can promote the hydration reaction of cement, thereby improving the early strength of CTB. Moreover, SO_4^{2-} in acidic solution reacts with $\text{Ca}(\text{OH})_2$ inside the backfill, and the product continuously fills the pores inside CTB, while OH^- in the alkaline environment can accelerate the cement's hydration response, thereby improving early CTB's strength. As time goes forward, the acid/alkaline corrosion extends to the interior of CTB, the hydration reaction continues, and there are more and more products, which lead to volume expansion and a large internal expansion force [33,34]. When exceeding a certain limit, a large number of cracks will be generated inside CTB, which will reduce CTB's compressive strength. The stronger the acid/alkaline environment is, the more obvious the effect of acid erosion/alkaline erosion, and the shorter time for the UCS to reach the peak [35].

3.2. Analysis of CTB Hydration Products

3.2.1. XRD Analysis

XRD is a rapid analysis technique used to identify the characteristics of crystalline substances. XRD analysis is used to track the differences in crystalline substances between CTB with various cement–sand ratios immersed in different solutions. Through XRD analysis, the difference of crystalline substances between CTB with various cement–sand ratios soaked in different pH solutions was tracked. The XRD diagram shows that CTB contains more crystalline and amorphous phases, including quartz, C-S-H, Ettringite (Aft), and $\text{Ca}(\text{OH})_2$. Quartz mainly comes from the original tailings, and C-S-H, Aft, and $\text{Ca}(\text{OH})_2$ are CTB's main hydration products. The characteristic peak of the hydrated calcium silicate gel appears near 22° and there is an obvious single diffraction peak. The $\text{Ca}(\text{OH})_2$ diffraction peak is near 16° , and there are obvious Aft diffraction peaks at 26° and 32° . From Figure 6, it can be seen that the peak strength of hydration products of CTB with cement–sand ratios of 1:10 and 1:15 changes significantly from 30 d to 60 days, and the number of hydration products is generated more during this period. When the cement-to-sand ratio was 1:20, the CTB's diffraction peak intensity changed little after 60 days, the hydration reaction proceeded slowly, and the total amount of hydration products was less. By 120 days, three different kinds of cement–sand ratio test cubes increased slightly in the diffraction peak count of CTB hydration products, the reaction tended to be stable, and the hydration was basically completed. At the same time, it is not difficult to see that the type of CTB hydration products with the same cement–sand ratio soaked in different pH solutions for different times is the same, but the quantity is different, and the increase in each time period is also different, which will affect the compressive strength of heavy metal elements in CTB and the heavy metal solidification ability of CTB [36].

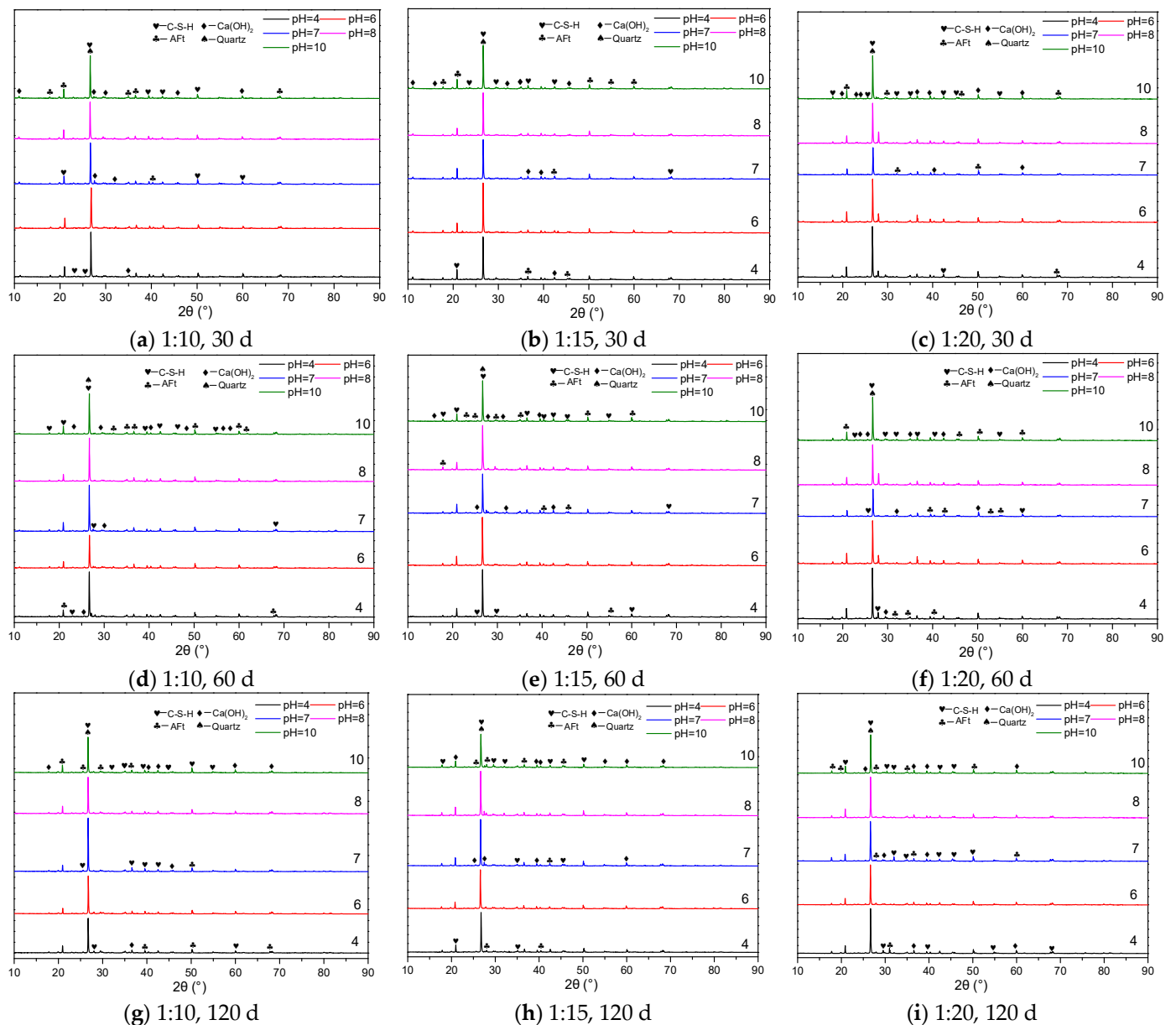


Figure 6. XRD diagrams of CTB with different cement–sand ratios immersed in a solution of pH = 4, 6, 7, 8, 10 for different days. (a) CTB with cement–sand ratio of 1:10 immersed in different pH solutions for 30 days. (b) CTB with cement–sand ratio of 1:10 immersed in different pH solutions for 30 days. (c) CTB with cement–sand ratio of 1:10 immersed in different pH solutions for 30 days. (d) CTB with cement–sand ratio of 1:10 immersed in different pH solutions for 60 days. (e) CTB with cement–sand ratio of 1:15 immersed in different pH solutions for 60 days. (f) CTB with cement–sand ratio of 1:20 immersed in different pH solutions for 60 days. (g) CTB with cement–sand ratio of 1:10 immersed in different pH solutions for 120 days. (h) CTB with cement–sand ratio of 1:15 immersed in different pH solutions for 120 days. (i) CTB with cement–sand ratio of 1:20 immersed in different pH solutions for 120 days.

According to the XRD diagram, it can be seen that the hydration product of CTB exhibits an increasing diffraction peak intensity during a soaking time of 120 days, which indicates that the hydration reaction continues and the hydration product is constantly generated. After soaking for 120 days, the peak strength of CTB hydration products with a cement–sand ratio of 1:10 in different soaking solutions is $\text{pH} = 7 > \text{pH} = 10 > \text{pH} = 6 > \text{pH} = 8 > \text{pH} = 4$, the peak strength of CTB hydration products with a cement–sand

ratio of 1:15 is $\text{pH} = 8 > \text{pH} = 7 > \text{pH} = 6 > \text{pH} = 4 > \text{pH} = 10$, and the peak strength of CTB hydration products with a cement–sand ratio of 1:20 is $\text{pH} = 4 > \text{pH} = 8 > \text{pH} = 6 > \text{pH} = 10 > \text{pH} = 7$. It can be concluded that during the long-term soaking process, CTB with higher cement content generates the most hydration products in a neutral environment. As the cement consumption decreases, the hydration products of CTB in acidic/alkaline environments gradually increase.

3.2.2. SEM Examination

SEM analysis was performed on CTB soaking for 30 d and 60 d in $\text{pH} = 4, 7, 10$ with three different cement–sand ratios and Figures 7 and 8 display the results. Aft, C-S-H, and $\text{Ca}(\text{OH})_2$ were found in the solidified structure of CTB. Aft's needle-shaped form serves as a supporting function for the skeleton. On the surface of CTB, C-S-H is covered with honeycomb to improve the mutual cementation of particles. The tight connections between Aft and C-S-H, as well as the cross-filling of the big pores between the particles, minimize porosity and form a physical package, which improves structural integrity and contributes to the macro-level strength of CTB. In addition, it was found that $\text{Ca}(\text{OH})_2$ was in the shape of a hexagonal sheet, which proved that CTB had a certain alkalinity [37,38].

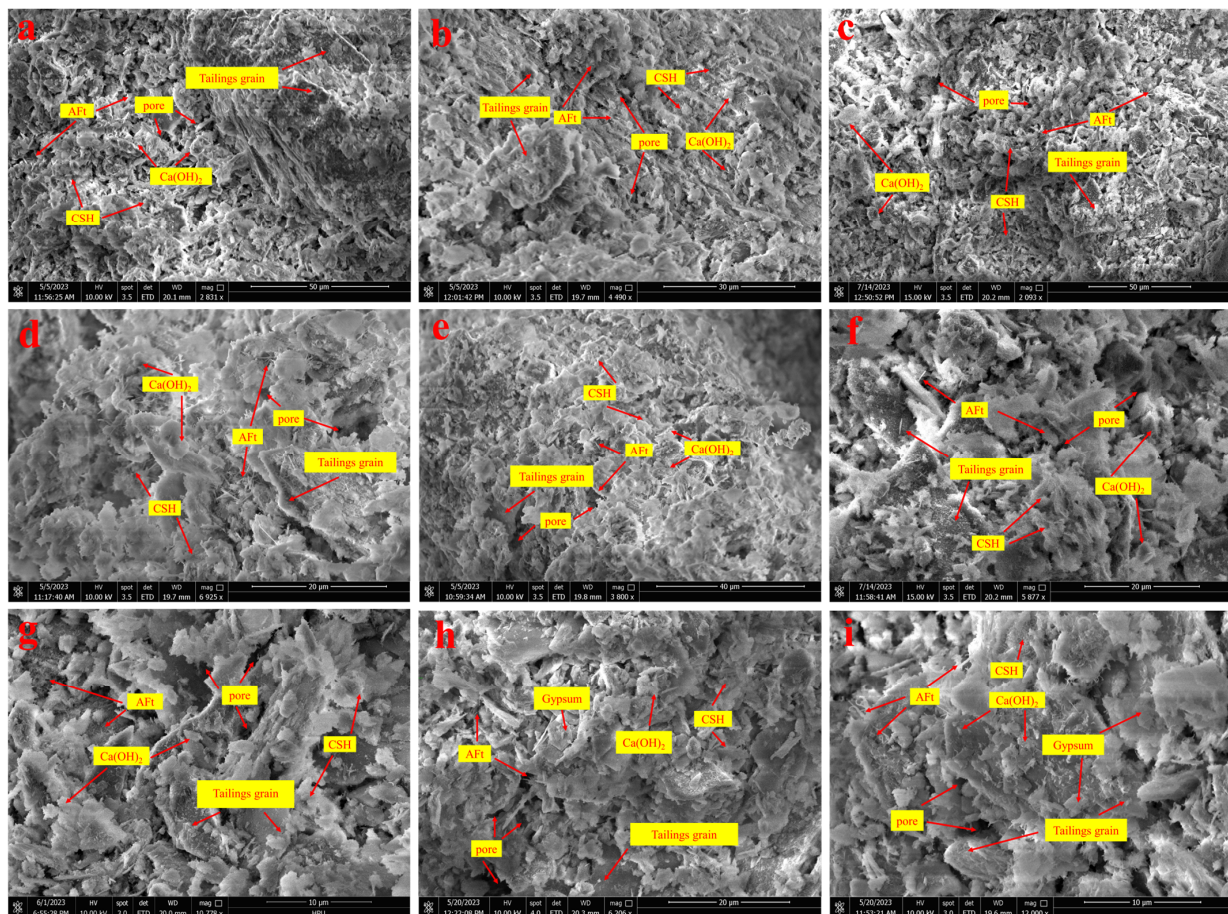


Figure 7. SEM image of CTB with cement–sand ratios of 1:10, 1:15, and 1:20 immersed in different solutions for 30 days. (a) CTB with cement–sand ratio of 1:10 in a solution of $\text{pH} = 4$. (b) CTB with cement–sand ratio of 1:10 in a solution of $\text{pH} = 7$. (c) CTB with cement–sand ratio of 1:10 in a solution of $\text{pH} = 10$. (d) CTB with cement–sand ratio of 1:15 in a solution of $\text{pH} = 4$. (e) CTB with cement–sand ratio of 1:15 in a solution of $\text{pH} = 7$. (f) CTB with cement–sand ratio of 1:15 in a solution of $\text{pH} = 10$. (g) CTB with cement–sand ratio of 1:20 in a solution of $\text{pH} = 4$. (h) CTB with cement–sand ratio of 1:20 in a solution of $\text{pH} = 7$. (i) CTB with cement–sand ratio of 1:20 in a solution of $\text{pH} = 10$.

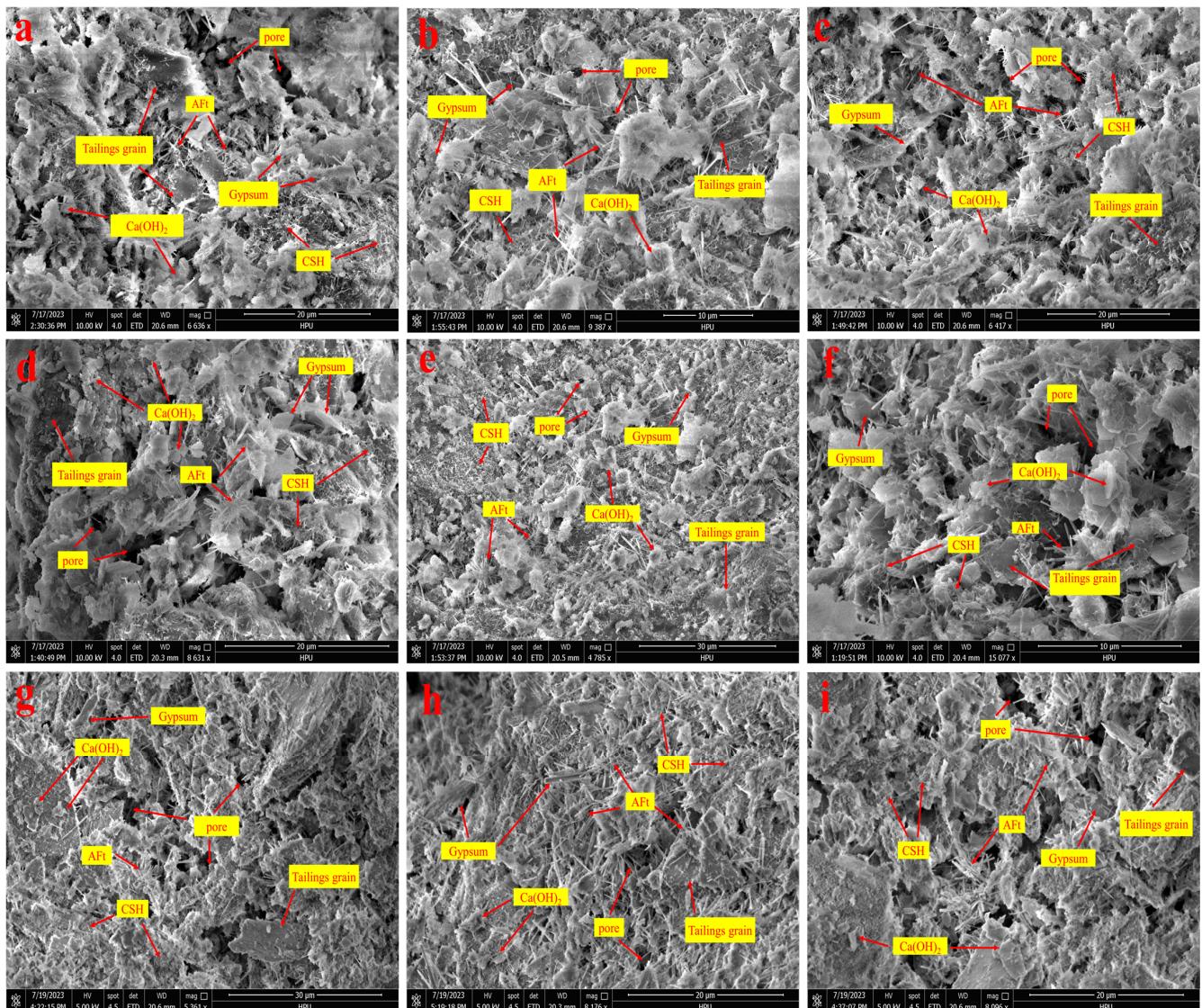


Figure 8. SEM image of CTB with different cement–sand ratio immersed in different solutions for 60 days. (a) CTB with cement–sand ratio of 1:10 in a solution of pH = 4. (b) CTB with cement–sand ratio of 1:10 in a solution of pH = 7. (c) CTB with cement–sand ratio of 1:10 in a solution of pH = 10. (d) CTB with cement–sand ratio of 1:15 in a solution of pH = 4. (e) CTB with cement–sand ratio of 1:15 in a solution of pH = 7. (f) CTB with cement–sand ratio of 1:15 in a solution of pH = 10. (g) CTB with cement–sand ratio of 1:20 in a solution of pH = 4. (h) CTB with cement–sand ratio of 1:20 in a solution of pH = 7. (i) CTB with cement–sand ratio of 1:20 in a solution of pH = 10.

Figure 7 illustrates how the soaking solution's pH increased over the course of 30 days. The hydration products increased and the pores were filled, which enhanced the structure of CTB. In the same acid/alkali environment, as the cement–sand ratio increases, the porosity decreases and the permeability of CTB decreases. This shows that the amount of cement is proportional to the strength of CTB, and in a certain range, CTB with a smaller cement–sand ratio is more conducive as the cement–sand ratio is more conducive to the rapid increase in CTB's strength in an alkaline environment. It can be seen from the comparison between Figures 7 and 8 that the hydration products in CTB increased greatly within 60 days of soaking. A large amount of Aft is accumulated and widely distributed. It is a needle-like crystal with a length of about 8–10 μm . It radiates from the edge of the pore to the inside of the pore in a cluster crystal form, and it partially transforms from needle-like to rod-like. At the same time, gypsum with a flat and coarse shape appeared, and the number of C-S-H

decreased significantly. In an acid/alkali environment, the gel's volume is reduced, and a large number of needle-like Aft intersects and overlaps with each other, making CTB loose and porous. The swelling effect significantly reduces the bond strength between the cementitious material and the tailings particles [39,40]. The macroscopic compressive strength of CTB decreases as a result of its increased diameter, increased number of pores, and ongoing crack growth. However, the increment of hydration products of CTB in a neutral environment is relatively small. The rise in Aft does not have expansibility and does not approach CTB's ultimate tensile strength. On the contrary, it will play a certain filling effect on the internal holes, and the generated hydration products and the inherent crystals of the raw materials are intertwined and bonded with each other, which increases the macroscopically strong CTB and gives the structure more density. At the same time, the compressive strength of CTB in the soaking solution with pH = 7 is the highest, which is in line with the actual measured compressive strength.

3.3. Leaching Characteristics and Solidification Mechanism of Heavy Metal Elements in CTB

Cement is an effective means to solidify/stabilize harmful substances. Products of hydration like C-S-H, Aft, and $\text{Ca}(\text{OH})_2$ are generated by a cement hydration reaction to accommodate heavy metals in physical and chemical forms and fix them in backfill, and they gradually harden into solidified bodies [41,42]. The solidification methods of heavy metal element include the physical encapsulation of dense hydration products, the physical adsorption of micropores of hydration products, the chemical fixation of a hydration reaction, a metathesis precipitation reaction, and isomorphous replacement [42,43]. Research on heavy metal elements' leaching properties and solidification mechanism can help control relevant variables, lessen heavy metal migration and volatilization, and lessen the likelihood that the ecological environment will be contaminated. In this paper, Cu, Zn, Pb, and Ni in CTB were selected for further study.

3.3.1. Cu

Figure 9 shows the cumulative leaching concentration of Cu element in specimens with three different cement–sand ratios soaked in different pH leaching solution for 120 days. Figure 9 shows that after a period of 10–60 days, the leaching concentration of Cu increased steadily in the specimens with the three varied cement–sand ratios, but the growth amount of each stage was different and gradually decreased. The capacity of CTB to solidify heavy metal ions keeps growing due to the ongoing hydration reaction and ongoing hydration products. The leaching rate of Cu element in the specimens with cement–sand ratios of 1:10 and 1:15 gradually stabilized in 60–90 d and 90–120 d, and the leaching concentration in the two stages showed a downward trend. However, the leaching rate of Cu element in specimens with a cement–sand ratio of 1:20 increased significantly, and the leaching amount showed a rising trend. At the same time, combined with the SEM diagrams (see Figures 7 and 8), the number of hydration products like C-S-H and Aft hardly changed during this period, with a small increase. It can be speculated that the decrease in cement content leads to the decrease in the hydration reaction ability and the shortening of reaction duration. In this time, the hydration reaction has been basically completed. With the continuous release of Cu^{2+} , the lattice capacity of the chemical reaction decreases, and the solidification ability of CTB to heavy metals decreases.

Cu element is easier to release in an acidic environment, and the cumulative leaching concentration is the lowest in a weak alkaline environment. In the environment of pH = 6–8, with the increase in pH, a $\text{Cu}(\text{OH})_2$ precipitate will be formed, and the concentration of Cu^{2+} leaching will decrease in turn. However, in the alkaline environment of the leaching solution pH = 10, Cu^{2+} mainly exist in the form of a hydrated phase ($\text{CuO} \cdot 3\text{H}_2\text{O}$), and Cu^{2+} will cooperate with Ca^{2+} to form a double hydroxide $\text{Ca}_2(\text{OH}) \cdot 4\text{Cu}(\text{OH})_2 \cdot 2\text{H}_2\text{O}$, thereby increasing the leaching concentration. Combined with the XRD diagrams, Figure 6 illustrates that CTB's hydration reaction during the test generates hydration products like C-S-H and Aft [44]. C-S-H is capable of absorbing a significant quantity of Cu^{2+} , resulting in

the formation of the C-S-H-Cu complex [45]. Cu^{2+} can also enter the columnar structure of Aft and replace Ca^{2+} by isomorphous substitution, thus being fixed, as shown in Figure 10.

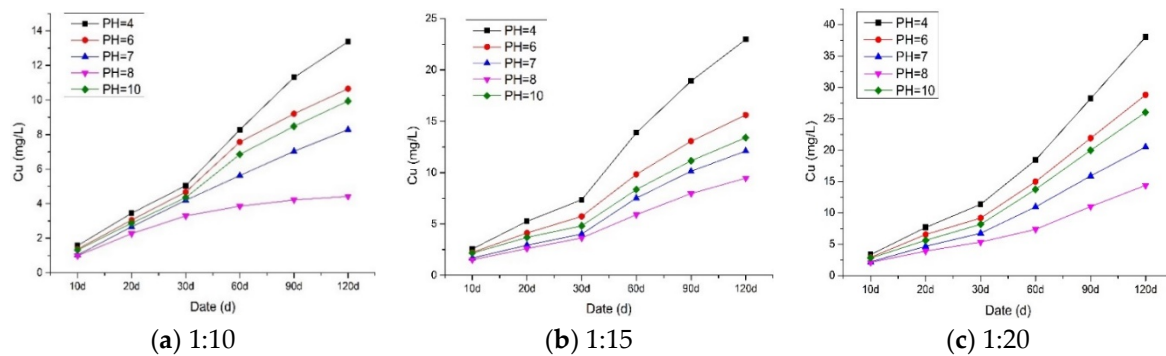


Figure 9. Cumulative leaching concentration of Cu^{2+} in CTB with different cement–sand ratios in different pH solutions. (a) CTB with cement–sand ratio of 1:10. (b) CTB with cement–sand ratio of 1:15. (c) CTB with cement–sand ratio of 1:20.

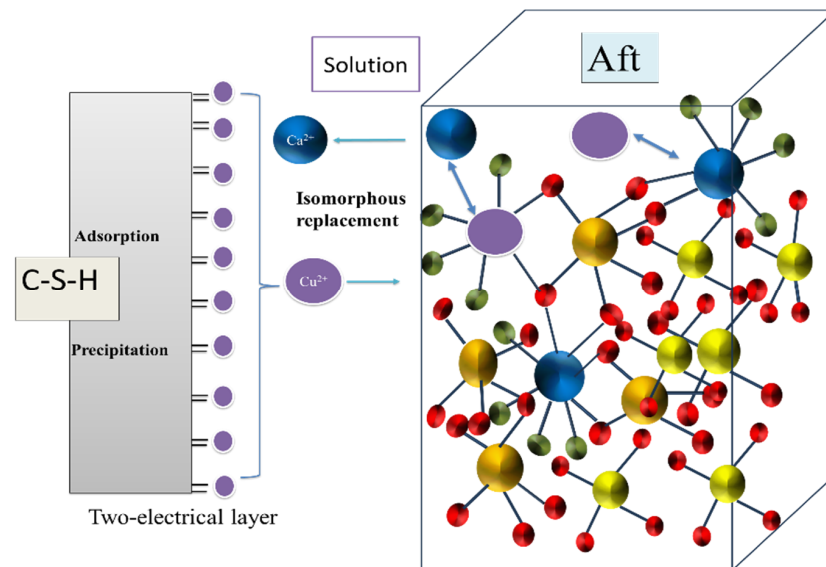


Figure 10. Solidification form of Cu. Adapted with permission from Ref. [43]. Copyright 2024 Elsevier.

3.3.2. Zn

Figure 11 shows that the cumulative concentration of Zn in CTB with different cement–sand ratios within 120 days of soaking in the same pH solution is $1:10 > 1:15 > 1:20$. Zn^{2+} can enter the columnar structure of Aft for isomorphous replacement to be fixed. Combined with XRD test results (Figure 6), Aft's diffraction peak intensity with three different cement–sand ratios within 120 days is also $1:10 > 1:15 > 1:20$, which is in line with the actual measured results. With the increase in cement content and the increase in the cement–sand ratio, the solidification ability of CTB is obviously improved, and the difference in the leaching concentration of Zn soaked in different environment for the same time becomes larger and larger. Within 0–30 days, with the initial progress of the hydration reaction, leaching per 10 days showed a trend of increasing first and then decreasing. After 30 days, the leaching amount of each stage decreased significantly, and the total leaching concentration tended to be stable. After 60 days, the cumulative leaching concentration of CTB with a cement–sand ratio of 1:20 showed a significant turning point, and there was an abrupt rise in the amount of heavy metal leaching. Combined with the SEM image (Figure 7), it can be seen that this is because the hydration reaction of CTB with various cement–sand ratios is the most intense within 30 days, and a large number of hydration

products are generated. After 30 days, the increment of hydration products gradually decreases in the same time period (Figure 8), and the increase in hydration products of CTB with a cement–sand ratio of 1:20 tends to be stable after 60 days. With the large release of heavy metals, the curing ability is relatively weakened, so the leaching amount of heavy metals increases significantly. When soaked for 120 days, the cumulative leaching concentration of Zn decreased with the increase of pH value in the range of pH = 4–10, and the maximum cumulative leaching concentration of CTB with a cement–sand ratio of 1:20 in the leaching solution of pH = 4 is 35.123 mg/mL, and the minimum is CTB with a cement–sand ratio of 1:10 in the leaching solution of pH = 10, being 4.869567 mg/mL.

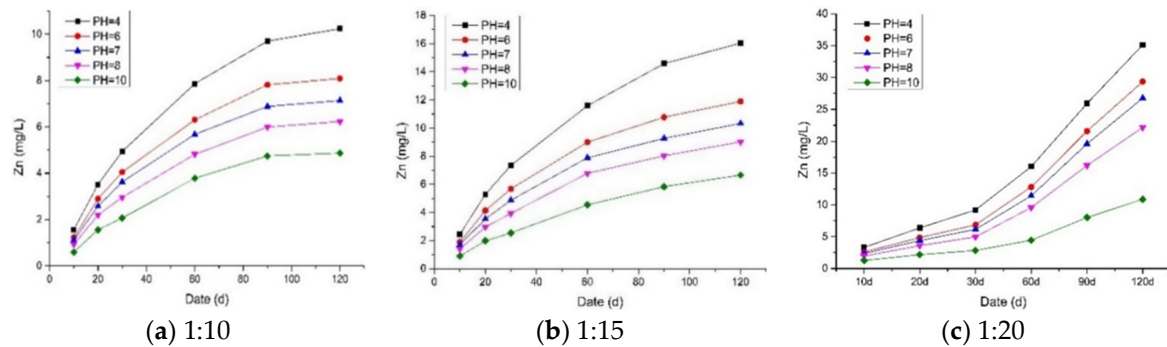


Figure 11. Cumulative leaching concentration of Zn^{2+} in CTB with different cement–sand ratios. (a) CTB with cement–sand ratio of 1:10. (b) CTB with cement–sand ratio of 1:15. (c) CTB with cement–sand ratio of 1:20.

When Zn^{2+} and OH^- react during cement hydration, an amorphous layer of $\text{Zn}(\text{OH})_2$ is formed in the solution, which is adsorbed on the surface of hydration products by chemical adsorption [46,47]. Therefore, Zn is easy to leach under acidic conditions, and the leaching amount changes, obviously. In the alkaline environment, zinc hydroxide exists in the form of $\text{Zn}(\text{OH})_4^{2-}$ and $\text{Zn}(\text{OH})_5^{3-}$, which can further react with Ca^{2+} to form a Ca–Zn hydrate precipitate $\text{Ca}[\text{Zn}(\text{OH})_3\text{H}_2\text{O}]_2$, which greatly reduces the amount of Zn^{2+} , as shown in Figure 12.

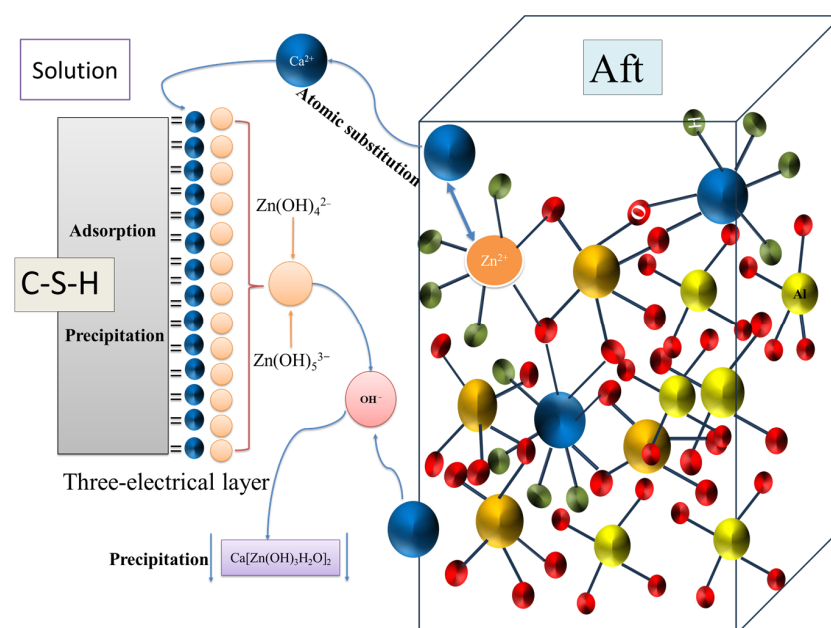


Figure 12. Solidification form of Zn. Adapted with permission from Ref. [43]. Copyright 2024 Elsevier.

3.3.3. Pb

Figure 13 illustrates the correlation between the cumulative leaching concentration of lead and soaking duration, the cement–sand ratio, and pH. For CTB soaked in the same pH solution, the cumulative leaching concentration of Pb in CTB with a small cement–sand ratio is always higher than that in CTB with a big cement–sand ratio as the soaking period increases. This is due to the high physical adsorption capability of C-S-H for Pb^{2+} . In addition, hydration products with a lot of structural spots on their surface where cations and anion might connect are C-S-H and Aft, which allow them to adsorb more heavy metal ions. Consequently, adsorption is crucial to Pb^{2+} 's solidification [48]. The total physical adsorption capability of Pb increases with the abundance of hydration products in CTB. Figure 14 depicts the form of Zn solidification.

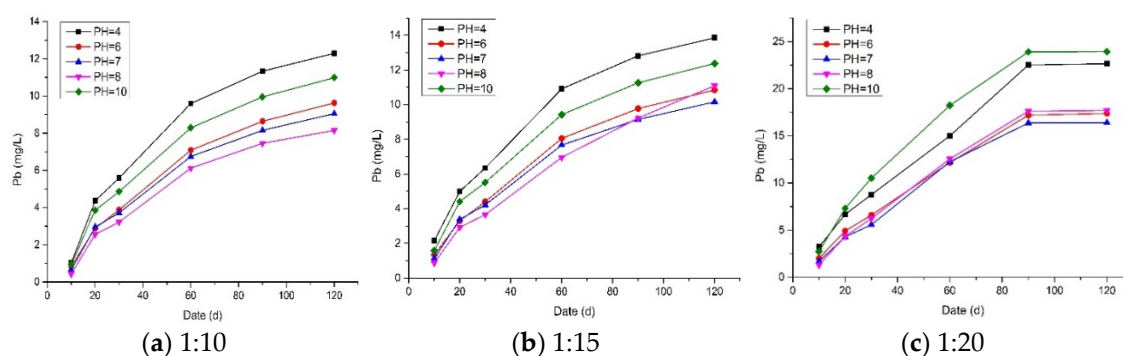


Figure 13. Cumulative leaching concentration of Pb^{2+} in CTB with different cement–sand ratios in different pH solutions. (a) CTB with cement–sand ratio of 1:10. (b) CTB with cement–sand ratio of 1:15. (c) CTB with cement–sand ratio of 1:20.

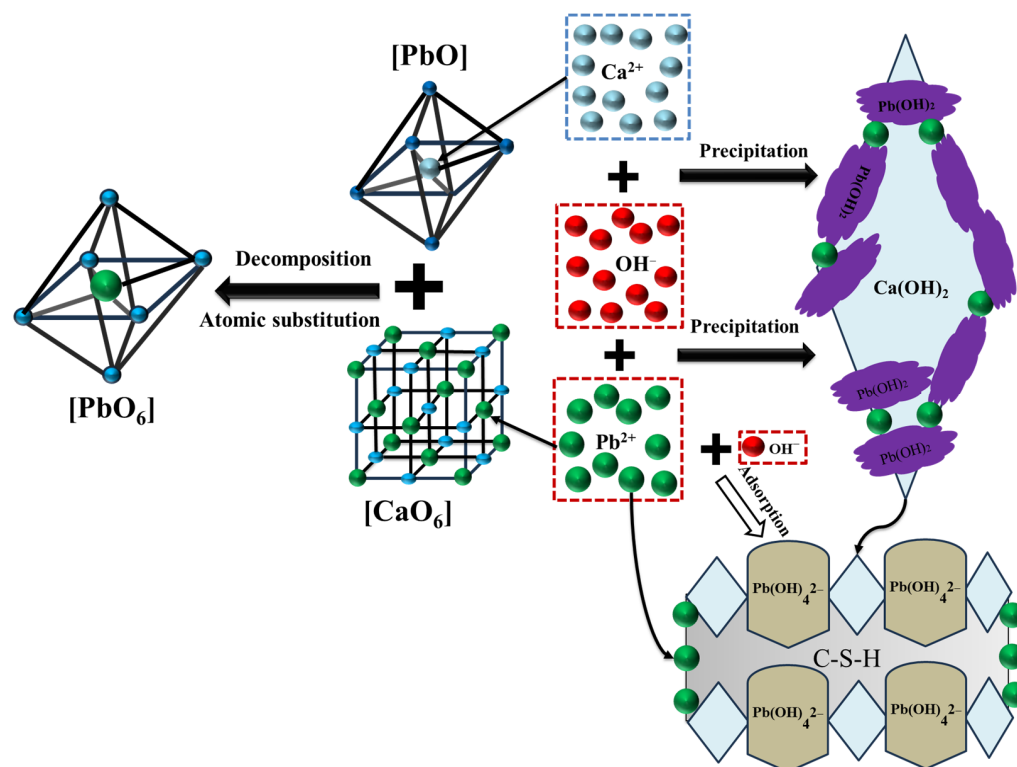
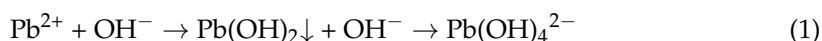


Figure 14. Solidification form of Pb^{2+} . Adapted with permission from Ref. [43]. Copyright 2024 Elsevier.

Figure 13a shows that, at a cement–sand ratio of 1:10, the order of Pb leaching concentration is $\text{pH} = 4 > 10 > 6 > 8 > 7$. $\text{Pb}(\text{OH})_2$ precipitates in an alkaline environment, while

$\text{Pb}(\text{OH})_2$, as zwitterionic hydroxides in an alkaline environment in the middle and late stages, will generate $\text{Pb}(\text{OH})_4^{2-}$, as shown in Equation (1). Under acidic conditions, the precipitation of Pb hydroxides and carbonates will be dissolved in the presence of Pb^{2+} . In an acidic environment, the precipitation of Pb hydroxides and carbonates will be dissolved by H^+ , thus existing as Pb^{2+} [49]. In a neutral environment, the precipitation of Pb is more stable. From Figure 13b, CTB with a cement–sand ratio of 1:15 is soaked for 120 days, and the cumulative leaching concentration in the solution with $\text{pH} = 8$ is higher than that in the solution with $\text{pH} = 6$ and $\text{pH} = 7$. The leaching has basically reached saturation at 90–120 days, and the leaching amount of Pb^{2+} is slightly less and tends to be stable [50]. However, in the weak alkaline environment of $\text{pH} = 8$, a part of the Pb precipitation was still dissolved, and the cumulative concentration increased slightly. Similarly, Figure 13c illustrates that this effect is more pronounced in CTB with a cement–sand ratio of 1:20, and a cumulative leaching concentration of $\text{pH} = 8$ solution at 120d is second only to $\text{pH} = 10$.



3.3.4. Ni

Figure 15 displays the total leaching amount of CTB after three distinct cement–sand ratios were soaked in various pH solutions for 120 days. It is evident that continuous leaching occurs at $\text{pH} = 4$ or $\text{pH} = 10$, where the leaching concentration of CTB is maximum. This is because Ni element exists in CTB in the form of $\text{Ni}(\text{OH})_2$, and $\text{Ni}(\text{OH})_2$ is an amphoteric compound. It can be precipitated in large quantities in acidic and alkaline environments. Under acidic conditions, $\text{Ni}(\text{OH})_2$ reacts with H^+ and transforms into Ni^{2+} in the solution [51]. The leaching behavior of Ni is controlled by the occurrence form of Ni element. The system's primary source of Ni is the amorphous precipitation $\text{Ni}(\text{OH})_2$. At the same time, Ni^{2+} replace Ca^{2+} in silicate minerals to form a substitutional solid solution, as shown in Figure 16.

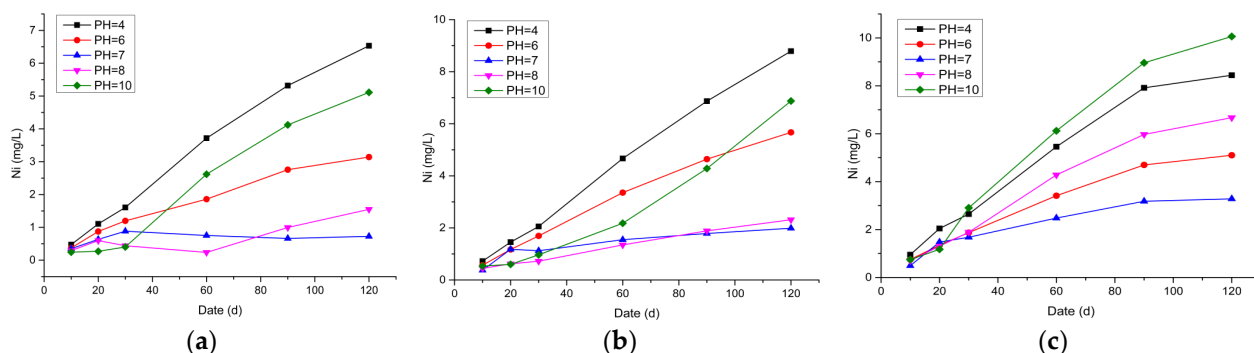


Figure 15. Cumulative leaching concentration of Ni^{2+} in CTB with different cement–sand ratios of 1:10, 1:15, 1:20 in different pH solutions. (a) CTB with cement–sand ratio of 1:10. (b) CTB with cement–sand ratio of 1:15. (c) CTB with cement–sand ratio of 1:20.

The amount of Ni^{2+} that leaches increases as pH decreases. Similarly, under the condition of a strong alkali, $\text{Ni}(\text{OH})_2$ in CTB reacts with OH^- in the solution to form HNiO_2^{2-} , and the leaching concentration rises as the pH value rises. Similarly, under the condition of a strong alkali, HNiO_2^{2-} is created when $\text{Ni}(\text{OH})_2$ in CTB combines with OH^- in the solution. The larger the pH value is, the more the cumulative leaching concentration is after soaking for the same time. In the alkaline solution of $\text{pH} = 8$ and $\text{pH} = 10$, the leaching concentration of Ni in CTB with a cement–sand ratio of 1:10 is less in 10–30 days, which is lower than that in other pH solutions, and the leaching concentration of CTB in other pH solutions is also less in this period. This is because a large amount of $\text{Ni}(\text{OH})_2$ precipitate is generated at the same time as the Ni^{2+} leaching, which is also the reason why other pH solutions have less leaching at the beginning. The leaching amount of CTB soaked in $\text{pH} = 7$ solution for 10–20 days and 20–30 days and the leaching amount of CTB

soaked in pH = 8 solution for 0–30 days and 30–60 days showed a downward trend. This is because during this time period, the solidification amount of Ni^{2+} by CTB began to be greater than the leaching amount, but with the decrease in hydration reaction efficiency, the solidification ability of CTB began to weaken, and then the leaching amount of Ni began to increase in the same time period. With the increase in the cement–sand ratio, the amount of tailings increases and the consumption of cement decreases, which leads to the weakening of CTB hydration ability, and the difference of the heavy metal leaching concentration between different pH leaching solutions becomes larger and larger. The cumulative leaching concentration of CTB with a cement–sand ratio of 1:20 in the solution with pH = 8 is second only to that in the solution with pH = 4 [52]. The leaching amount of 10–20 days in neutral environment is slightly larger than that in the weak acid and weak alkali solution, but the cumulative leaching concentration is finally the smallest in different pH solutions.

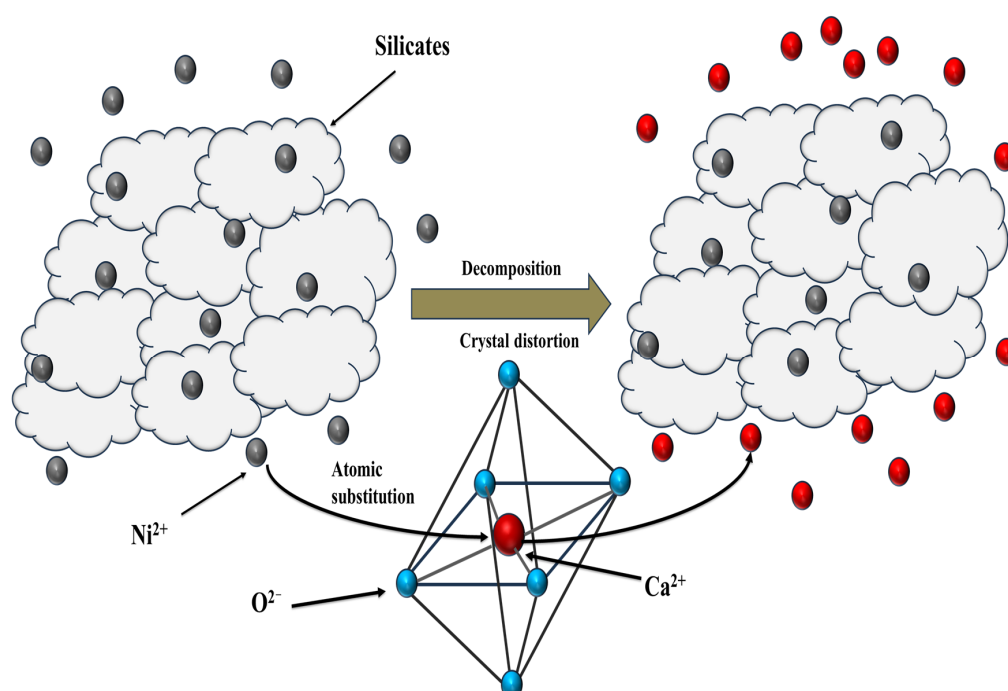


Figure 16. Solidification form of Ni^{2+} . Adapted with permission from Ref. [43]. Copyright 2024 Elsevier.

3.4. Kinetics of Leaching

3.4.1. First-Order Model of Reaction and Diffusion (FRDM)

Leaching behavior is usually dominated by dissolution and diffusion. The first-order reaction model (FRM) involves the dissolution of matter at the interface between solid and liquid (Equations (2) and (3)). The bulk diffusion model (BDM) takes into account the transfer of dissolved matter in porous media (Equations (4) and (5)). The first order reaction/diffusion model (FRDM) can be used to study the leaching of elements in specimens through a simplified mass transfer mechanism (Equation (6)). It can more accurately depict how heavy metals in CTB leach and calculate the accumulation of leaching pollutants. Suzuki, Ahmari, and Zhang et al.'s model also proved that the model is accurate enough for the leaching kinetics of solid media, indicating the correlation between the dissolution rate of elements and the total amount of soluble elements in the specimens, and the diffusion of elements follows Fick's law [53–55].

$$\frac{dQ}{dt} = -kQ, \quad Q|_{t=0} = Q_0 \quad (2)$$

Q : the amount of soluble elements;

K : rate constant;

Q : initial amount of soluble metals.

$$M = Q_0[1 - \exp(-kt)] \quad (3)$$

M : element accumulation.

$$\frac{\partial C}{\partial t} = D_e \frac{\partial^2 C}{\partial x^2} \quad (4)$$

C : measurement concentration at intervals;

D_e : effective coefficient;

X : direction.

$$M = 2AC_0\sqrt{\frac{D_e t}{\pi}} \quad (5)$$

A : surface area;

C_0 : total concentration of the contaminant in the solid specimen.

$$M = Q_0[1 - \exp(-kt)] + 2AC_0\sqrt{\frac{D_e t}{\pi}} \quad (6)$$

The effective diffusion coefficient (D_e), which only shows the impact of tortuous pores and their connection on pollutant transport, can be utilized to distinguish between physical and chemical retardation.

3.4.2. Evaluation of Heavy Metal Leaching Kinetics

The nonlinear regression method of Microsoft Excel of Office16 solver is used to fit the measured heavy metal leaching concentration and time data to the FRDM. The model parameters are obtained by nonlinear regression analysis under the minimum mean square error, so as to evaluate the leaching kinetics. It can be seen from the above research results that CTB with a cement–sand ratio of 1:15 basically meets the requirements of environmental and structural safety. Therefore, the specimens with a cement–sand ratio of 1:15 were selected to calculate the reverse calculation parameters of four heavy metals in different pH solutions. Table 6 displays the computation results, and Figure 17 displays the fitting curve. It is evident that the experimental data is well fit by Equation (6).

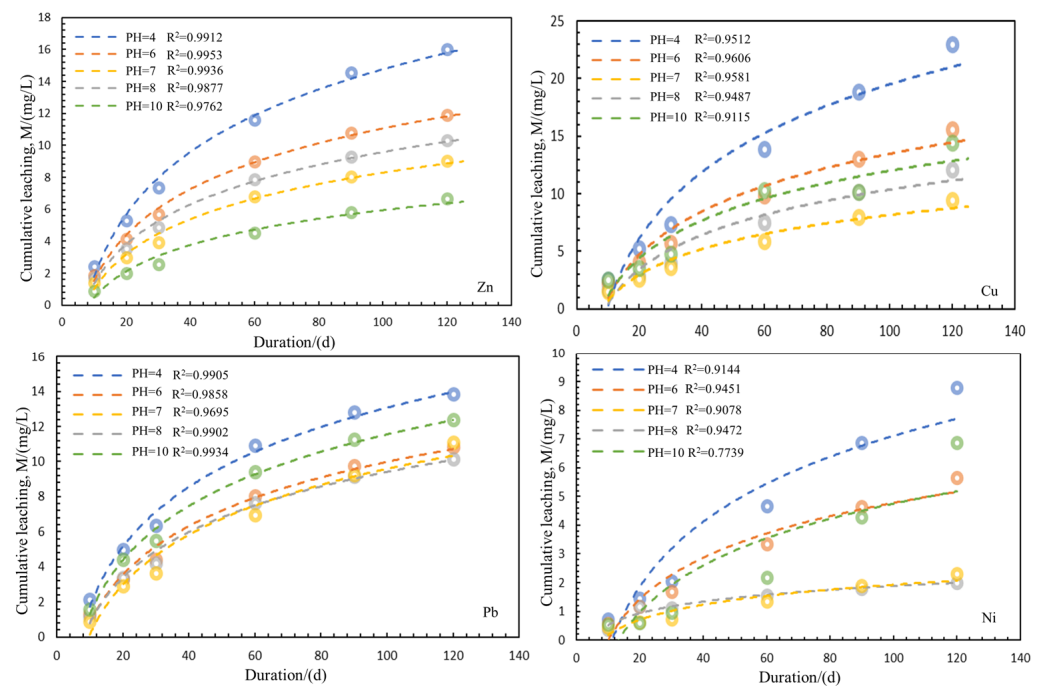


Figure 17. FRDM predicts concentration of Zn, Cu, Pb, Ni.

Table 6. Summary of back-predicted FRDM parameters for Cu, Zn, Pb, and Ni.

C/S	Element	Solutions	A(cm ²)	Q ₀ (mg/mL)	k (1/h)	D _e (cm ² /h)
1:15	Zn	pH = 4	49.98	1.058159	1.977422	2.1×10^{-5}
		pH = 6	49.98	0.925473	1.958337	1.47×10^{-5}
		pH = 7	49.98	0.870359	1.943597	1.22×10^{-5}
		pH = 8	49.98	0.794895	1.918844	1.01×10^{-5}
		pH = 10	49.98	0.654149	1.838956	6.35×10^{-6}
	Cu	pH = 4	49.98	1.167262	0.820333	2.95×10^{-5}
		pH = 6	49.98	1.007619	0.66165	1.89×10^{-5}
		pH = 7	49.98	0.882957	0.320734	1.37×10^{-5}
		pH = 8	49.98	0.780359	0.062625	9.92×10^{-6}
		pH = 10	49.98	0.954293	0.592445	1.65×10^{-5}
	Pb	pH = 4	49.98	0.544189	0.992408	8.53×10^{-5}
		pH = 6	49.98	0.446199	0.866262	6.43×10^{-5}
		pH = 7	49.98	0.427495	0.748257	6.04×10^{-5}
		pH = 8	49.98	0.427731	0.499674	6.23×10^{-5}
		pH = 10	49.98	0.500768	0.944952	7.49×10^{-5}
	Ni	pH = 4	49.98	0.551586	0.820631	4.96×10^{-6}
		pH = 6	49.98	0.440933	0.724281	3.58×10^{-6}
		pH = 7	49.98	0.241322	0.479181	1.53×10^{-6}
		pH = 8	49.98	0.363111	0.642234	2.75×10^{-6}
		pH = 10	49.98	0.493019	0.770067	4.17×10^{-6}

Table 6 illustrates how the fitting function has a high coefficient of determination for each of the chosen elements submerged in various solutions. The effective diffusion coefficient D_e changes significantly with the cement–sand ratio, heavy metal elements, and reagent solution, demonstrating that chemical retardation is an important factor affecting the leaching behavior of CTB. The K value of Cu, Zn, Pb, and Ni in the specimens with a cement–sand ratio of 1:15 soaked in a neutral solution is low, indicating that it leaches poorly and reacts with the solution at the slowest rate. The leaching amount of the four elements in the leaching test is less under this environmental condition. The Q_0 values of Cu and Zn in an acidic environment, as well as Pb and Ni in an alkaline environment, are significantly higher than those in other environments, and the Q_0 value in CTB with less cement consumption is higher, demonstrating that each metal has a significant amount of soluble leftover components in the appropriate environmental circumstances. It is evident that the Q_0 values of Pb and Ni in an alkaline environment and Cu and Zn in an acidic environment are higher than those in other settings. The smaller the cement–sand ratio is, the larger the Q_0 value is in CTB, indicating that there is a large number of soluble residual elements in each metal under the corresponding environmental conditions. Zinc’s leaching kinetics are driven by reaction because it has a higher reaction rate and chemical retardation than those of other heavy metal elements. This poorer immobilization efficiency is a result of Zinc’s higher reaction rate [56].

4. Conclusions

In conclusion, the following conclusions are made in light of the research findings on the compressive strength of three distinct types of lime–sand ratio specimens in five various pH solutions, as well as the leaching properties and solidification ability of four heavy metals.

1. The decrease in cement consumption reduces the amount of hydration products of CTB, cement products such as hydrated calcium silicate decrease, the compactness of CTB decreases, the pores increase, and it is more vulnerable to damage and thus degraded, leading to a decrease in durability. At the same time, the solidification structure is reduced, which leads to the decrease in the heavy metal solidification ability of CTB, the increase in the heavy metal diffusion channel, and the increase in the heavy metal leaching risk.
2. In the stronger acid/alkaline environment, the effect of acid/alkaline erosion is more obvious, which will accelerate the dissolution of hydration products in CTB, reduce the long-term strength of CTB, and reduce the durability of CTB. In addition, when cracks increase, the permeability of heavy metals increases and the migration ability of heavy metals increases.
3. With regard to the typical heavy metals Cu, Zn, Pb, and Ni that this paper examines, it is found that the solidification methods include the physical encapsulation of dense hydration products, the physical adsorption of micropores of hydration products, the chemical fixation of hydration reaction, the metathesis precipitation reaction, and the isomorphous replacement reaction.
4. In a neutral environment with a pH of 7, the compressive strength and heavy metal solidification impact of CTB with a cement–sand ratio of 1:15 essentially satisfy the requirements for environmental and structural safety.
5. A satisfactory description of the heavy metals' leaching in low-consumption cement backfill can be obtained using the first-order reaction/diffusion model. This suggests that a key element influencing leaching behavior control is solubility or response rate.

In the future, the range of critical values for cement consumption will be further narrowed, and the mechanism of heavy metal ion infiltration and leaching will be explored in depth. At the same time, the strength of CTB will be controlled by changing process parameters and other methods, and it will be applied to mining practice.

Environmental Implication

Cement consumption is the key factor affecting the strength of the backfills and the solidification effect of heavy metal ions. Previous studies only focused on the strength of backfills under the influence of cement consumption, ignoring the environmental risk of heavy metal ion leaching. This study reveals the mechanism of low-strength backfill enhancement and the weak solidification of heavy metals from the perspective of chemistry and mechanics, and it studies the mechanical behavior and solidification mechanism of low-strength backfill under erosion, in order to break through the technical and economic bottleneck restricting the development of filling technology.

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