

Research Article

Comparative Experimental Study on Calcium Oxide, Calcium Hydroxide, and Calcium Carbonate Solidified Zinc-Contaminated Red Clay

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Calcium oxide is a common curing agent used for curing contaminated soils. It is often used with cement to adjust the pH value. Calcium carbonate and calcium hydroxide are common products of calcium oxide. The effects of three kinds of binder in curing Zn²⁺-contaminated red clay are compared. The Zn²⁺ concentrations were selected as 0.1 and 0.5%, and the curing agent was added separately. The values of the unconfined strength of soil samples were compared after the three curing agents were added to the soil alone. It was found that the strength of the specimen with the addition of calcium carbonate was lower than the strength of the contaminated soil without the addition of the curing agent, while the strength of calcium hydroxide and calcium oxide was enhanced with the addition of calcium hydroxide, in general, and the strength of calcium oxide was higher. When CaO is added, the strength of the sample with a 0.1% concentration of zinc-contaminated soil is greater than the strength of the sample with a 0.5% concentration. But when Ca(OH)₂ is added, the strength of the sample with 0.1% zinc-contaminated soil is less than the strength of the sample with a 0.5% concentration. Ca(OH)₂ provides enough OH⁻, and there are enough zinc ions to react with it in the high-concentration Zn²⁺-contaminated soil to form CaZn₂(OH)₆·2H₂O, which enhances the strength of the soil. By comparing the pH values of calcium oxide and calcium hydroxide, the pH of the soil is about 12, and the pH of calcium carbonate is about 8.

1. Introduction

Red clay is a kind of regional special soil, which is characterized by high water content, high porosity ratio, high plasticity, high strength, medium and low compressibility, and low permeability [1]. It is mainly distributed in Guangdong, Guangxi, Yunnan, Guizhou, and Sichuan [2] and is widely distributed in south China, especially in Guangxi. Guangxi is rich in mineral resources, and as the exploitation of resources has brought rapid development to Guangxi, it has also caused heavy metal pollution. In geotechnical engineering, many factors cause engineering accidents, including changes in rock joint surfaces, seepage pressure, water pressure, environmental conditions, and other causes that lead to changes in mechanical properties and engineering accidents.

Different particle shapes and different mechanical properties of the material are reported in the previous studies [3–8]. After the soil is contaminated, the soil particles dissolve, and the cementation strength is destroyed, thus making the mechanical properties of soil compressibility, shear strength, bearing capacity, etc. weakened, which can lead to a series of engineering accidents [9]. The mechanical strength of the soil samples was very significantly affected by the concentration of heavy metals [10, 11]. Some scholars investigated the Guangxi Daxin manganese mining area and the Sixing lead-zinc mining area, and they found that the zinc pollution in and around this area is relatively serious [12, 13]. The solidification/stabilization (S/S) technology is commonly used in the treatment of contaminated soil. S/S refers to the use of physical or chemical methods to fix pollutants in the soil

or to convert pollutants into chemically inactive forms to prevent them from migrating and spreading in the environment, thereby reducing the poisoning of pollutants. The degree of repair technology [14–17] has the advantages of relatively low cost, high strength of the treated foundation soil, and convenient construction [18]. The most widely used materials in S/S are cement and lime. Lime is an alkaline nonhydraulic cementitious material. Its effect on heavy metals in the soil is mainly to increase the pH of the soil and promote the precipitation of heavy metals into carbonates, silicates, and hydroxides [17, 19].

Chen et al. [20] found that heavy metal Zn^{2+} had obvious damage behavior to Guilin red clay through leaching test. Li et al. [21] found that Zn^{2+} changed the mineral composition of red clay and reduced the content of clay particles. The complex formed with amorphous free iron oxide and the iron oxide film on the surface of the pellets mutually repel each other, and the zinc salt produced destroys the soil particles. The structure between the red clay increases the pore volume and loosens the structure, reduces the interaction force between the soil particles, and leads to the attenuation of the macromechanical properties of the red clay remolded by Zn^{2+} in Guilin. Du et al. [22] found that the pore space increased with the increase of zinc ion concentration and the hydration products of cement decreased with the increase of cement curing zinc-contaminated kaolin. Reddy et al. [23] cured zinc ion-contaminated soil with new ternary blend limestone calcined clay cement (LC3) and found that the unconfined strength was greater when using the LC3 curing agent than when using cement. Xi and Xiong [24] added lime to cement to solidify Zn^{2+} -contaminated soil, which is much better than that of cement alone, indicating that lime has a better solidification effect on Zn^{2+} -contaminated soil.

$CaCO_3$ and $Ca(OH)_2$ are two common products of CaO in chemical reactions. This article compares the solidification of Zn^{2+} -contaminated red clay by $CaCO_3$, $Ca(OH)_2$, and CaO. The unconfined strength of each specimen after adding the curing agent was compared. The pH value of the specimens was tested. The specimen composition was analyzed using the XRD test, and the curing mechanism of the curing agent was speculated, to explore the curing effect of three binders on Zn^{2+} -contaminated red clay.

2. Materials and Methods

2.1. Test Materials. The experimental soil was selected from a construction site in Yanshan Town, Guilin City, China. The soil was brownish red. The red clay is air-dried, crushed, and passed through a 2 mm soil sieve. The basic physical properties of red clay are shown in Table 1.

In this experiment, $Zn(NO_3)_2 \cdot 6H_2O$ was used as the pollution source because NO_3^- has better solubility [25]. According to the paper [26], the pollution concentration is set to 0.1% and 0.5% (the percentage of the mass of Zn^{2+} to the mass of dryness) for the convenience of identification, denoting 0.1% and 0.5% of Zn^{2+} as Z1 and Z5. $CaCO_3$, $Ca(OH)_2$, and CaO are, respectively, marked as C, H, and O (for example, if the pollution concentration is 0.1%, the

content of CaO is 10%, and it is recorded as Z1O10), as shown in Table 2.

2.2. Sample Preparation and Test Method. The zinc nitrate solution was prepared with distilled water, the solution was mixed with red clay, and it was stirred evenly to make the contaminated soil with a water content of 30%. A certain concentration of curing agent was added to the contaminated soil and stirred, and 30% of the water was added to keep the initial water content of the contaminated soil at 30% after adding the curing agent. The soil was sealed for 12 hours, and its moisture content was measured. According to the Standard for Geotechnical Test Methods, the remolded lateral limit-free specimens with a height of 80 mm, a diameter of 39.1 mm, and a dry density of $1.4 g cm^{-3}$ were prepared by the striking sample method. The samples were cured in the curing box under standard conditions for one week. The test was carried out using a DW-1-type unconfined pressure instrument. The damaged samples were air-dried through a 2 mm sieve, 10 g, with the addition of pure water 50 ml (soil to water ratio of 1 : 5). They were shaken on an oscillator for 3 min and rested for 30 min for testing using the PHS-3C instrument. Some of the destroyed soil samples were then dried through a 0.075 mm sieve for XRD analysis.

3. Experimental Results and Analysis

3.1. Comparison with Curing Agent Stress-Strain Curve. Figure 1(a) shows the stress-strain curves of the samples after the addition of calcium carbonate. It can be seen from the figure that the strength of the sample is the highest when the Zn^{2+} concentration is 0.1% and the calcium carbonate content is 5% (i.e., Z1C5). This is followed by Z5C5, Z1C10, Z5C10, and finally Z1C15 and Z5C15, but the highest strength is 74.865 kPa and the lowest one is 30.30 kPa, which are relatively low values. When the content of calcium carbonate is 5%, the modulus (ratio of stress to strain) does not change much in the stress rise phase, while the modulus starts to become smaller in the stress fall phase; but when the content of calcium carbonate is greater than 10%, the modulus changes more in the stress rise and fall phase, both decreasing. At the same contamination concentration, the strength of the sample decreased with the increase of calcium carbonate content, indicating that $CaCO_3$ has a weakening effect on Zn^{2+} -contaminated red clay. The stress-strain curves of the specimens with the same dose of calcium carbonate are more similar.

Figure 1(b) shows the stress-strain curves of the samples after the addition of calcium hydroxide. The strength of the samples increased after the addition of calcium hydroxide. With the increase of the content, the strength of the specimens increased significantly. The specimen with the greatest strength was Z5H15, followed by Z5H10, Z1H15, Z1H5, Z5H10, and finally Z1H5. The maximum value was 1008.90 kPa, and the minimum value was 470.58 kPa. The failure strain decreased with the increase of the calcium hydroxide content. When the contamination concentration is smaller and the content of calcium hydroxide is less, the

TABLE 1: Basic physical properties of red clay.

Liquid limit (%)	Plastic limit (%)	Plasticity index	Optimum moisture content (%)	Maximum dry density (g cm^{-3})	Specific gravity	pH
50.70	33.18	17.52	30.78	1.53	2.72	4.67

TABLE 2: Basic physical properties of red clay.

	Zn^{2+}	CaCO_3	$\text{Ca}(\text{OH})_2$	CaO
1	0.1%	5%	5%	5%
2	0.5%	10%	10%	10%
3		15%	15%	15%

failure strain is larger, the Z5C15 failure strain is the smallest, and the Z1C5 failure strain is the largest. At a zinc ion concentration of 0.1%, the axial deformation of the samples was greater than that of the ion concentration of 0.5%. Most of the curves had small modulus values before the axial strain was 1%, indicating that the axial strain to about 1% was the initial loading stage of the stress-strain curve, which was a relatively gentle straight line, followed by a nonlinear rising stage of stress-strain. The initial loading stage is the specimen compacting stage, and the strain at the end of each specimen compacting is close, which is because the water content of the specimens mixed with calcium hydroxide is close, and their porosity is also similar. When the strength of the soil sample is larger, the curve of the nonlinear rising stage is steeper, and the axial strain value of this stage is smaller. It indicates that the sample becomes brittle material after adding calcium hydroxide, and the more obvious brittleness of the soil shows. After adding calcium hydroxide, the stress-strain curve of the samples with 0.5% contamination concentration is steeper than that of the samples with 0.1% contamination concentration. It indicates that the sample becomes a brittle material after adding calcium hydroxide, and the more obvious brittleness of the soil shows. After adding calcium hydroxide, the stress-strain curve of the sample with 0.5% contamination concentration is steeper than that of the specimen with 0.1% contamination concentration. It indicates that the brittle performance of the samples with a 0.5% contamination concentration is more obvious after the addition of calcium hydroxide.

When the concentration of calcium hydroxide is the same, the two contamination concentrations of the specimens are compared, and the sample with a 0.5% contamination concentration has a higher unconfined strength. This may be because the contaminated soil with 0.5% Zn^{2+} concentration provided more Zn ions, which allowed the Zn^{2+} to fully react with calcium hydroxide to produce $\text{CaZn}_2(\text{OH})_6 \cdot 2\text{H}_2\text{O}$, and $\text{CaZn}_2(\text{OH})_6 \cdot 2\text{H}_2\text{O}$ is insoluble and adds strength to the specimen. The reaction equation is as follows:

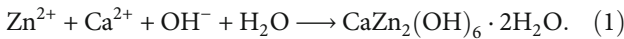


Figure 1(c) shows the stress-strain curves measured for the samples after the addition of calcium oxide. The highest

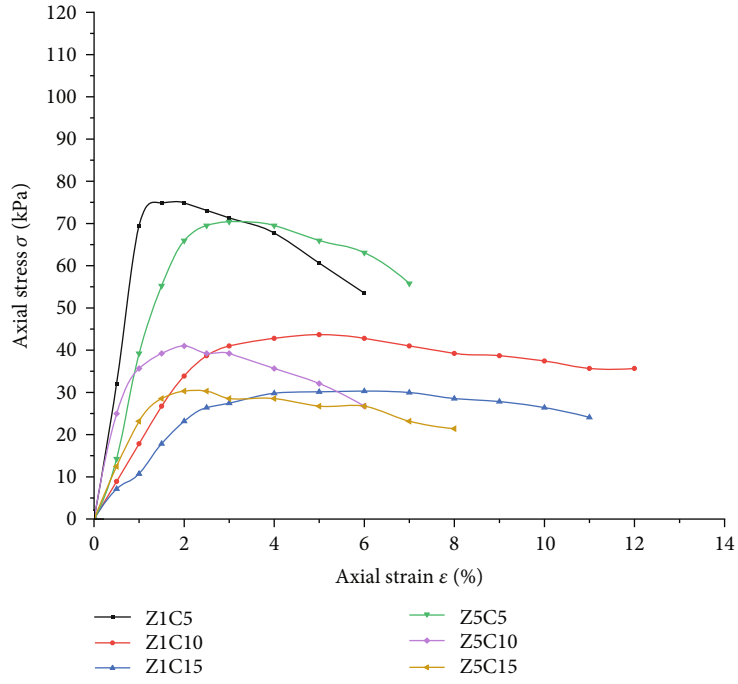
strength was Z1O15, followed by Z5O15, Z1O10, Z5O10, Z1O5, and finally Z5O5. The highest strength was 1328.14 kPa, and the lowest strength was 541.88 kPa. The strength of Z1O15 was twice as strong as that of Z1O5; the strength of Z5O15 was 2.3 times as strong as that of Z5O5. The stress-strain curves were similar for different contamination concentrations with the same calcium oxide content; the damage strain of 0.5% contaminated specimens was larger than that of 0.1% contaminated specimens, and with the increase of calcium oxide content, the curve of the nonlinear rising stage of 0.5% contaminated specimens was steeper than that of 0.1% contaminated specimens. The destructive strain gradually increased, the value of the destructive strain increased accordingly, and the initial loading phase became longer. The samples also became brittle after the addition of calcium oxide, while the brittleness of the specimens slightly diminished with the increase of calcium oxide content.

It can be found that with the addition of calcium oxide, the unconfined strength of the samples increased with the increase of the content. However, the strength of calcium oxide specimens with the same content weakened with the increase in contamination concentration. It indicates that the curing effect of calcium oxide will be weakened for high concentrations of contaminated soil. The strength of the samples with 10% calcium oxide was significantly higher than that of the specimens with 5% calcium oxide; however, the strength of the specimens with 15% calcium oxide was not much higher than that of the specimens with 10% calcium oxide.

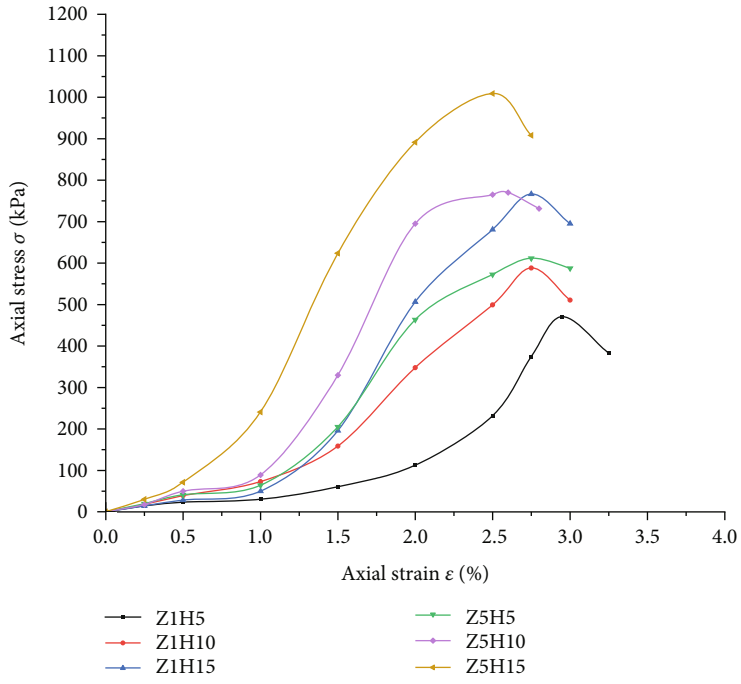
Calcium oxide is a cement-like cementitious material, and red clay is rich in SiO_2 , Al_2O_3 , and Fe_2O_3 , which is added to the soil for hydration reaction. It produces C-S-H, C-A-H, and calcium hydroxide, which have a good cementing effect and have an obvious enhancement effect on the strength of the soil.

3.2. The Unconfined Compressive Strength of Samples.

Figures 2 and 3 show the unconfined compressive strength at each curing agent content. The strength of the samples with the addition of calcium carbonate did not increase; on the contrary, the strength of the samples decreased with the increase of calcium carbonate admixture. According to Wang et al. [27], calcium carbonate was found to have a weakening effect on the red clay, which shows that calcium carbonate still has a weakening effect on the Zn^{2+} -contaminated red clay. The strength of contaminated clay with the addition of calcium oxide and calcium hydroxide was significantly higher than calcium carbonate. The strength of Z5H5 is higher than the compressive strength of Z5O5 as can be seen in Figure 3; however, with the increase in the amount of curing agent, the strength of the samples with the addition



(a)



(b)

FIGURE 1: Continued.

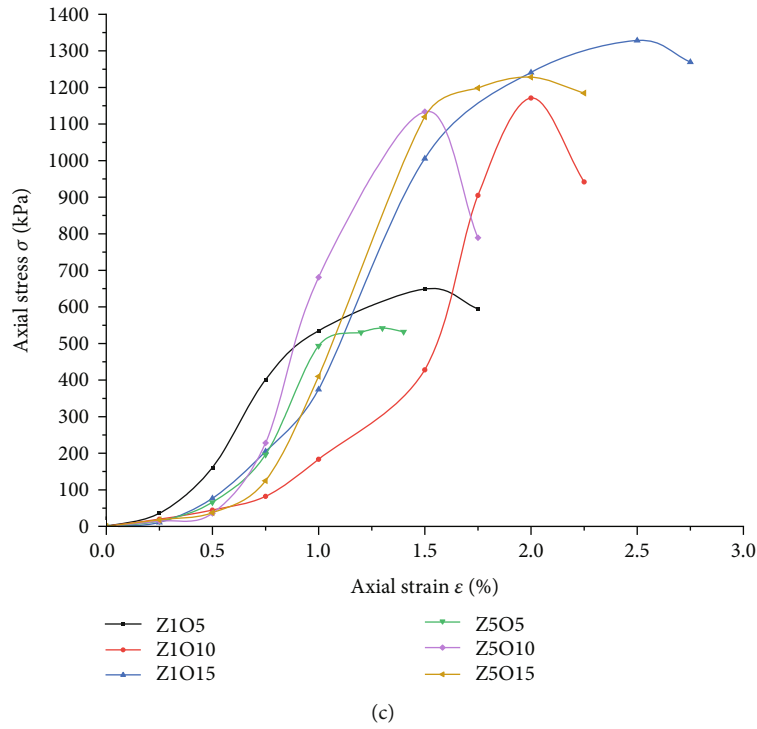


FIGURE 1: Stress-strain curves of each curing agent. (a) Stress-strain curve of the samples after adding calcium carbonate. (b) Stress-strain curve of the samples after adding calcium hydroxide. (c) Stress-strain curve of the samples after adding calcium oxide.

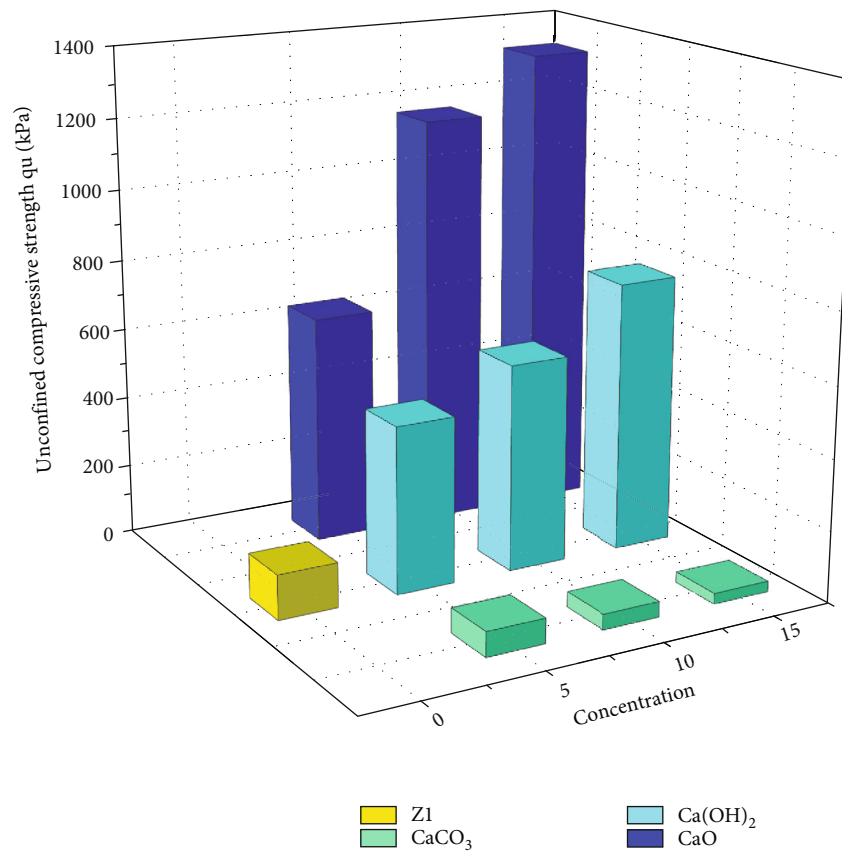


FIGURE 2: The unconfined compressive strength of each sample at 0.1% Zn^{2+} concentration.

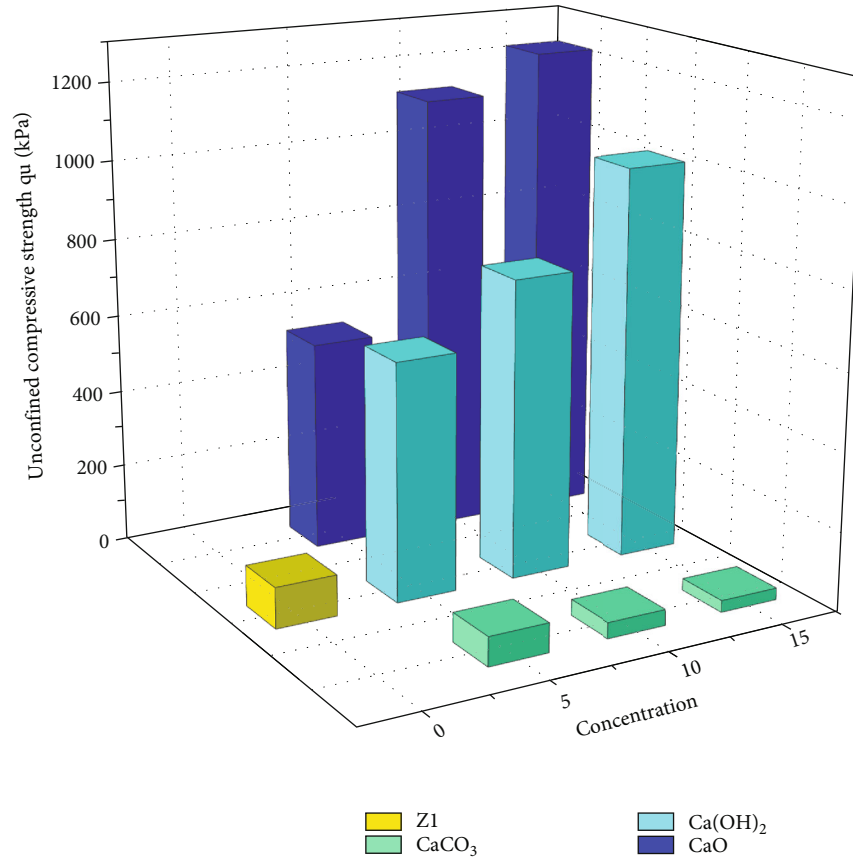


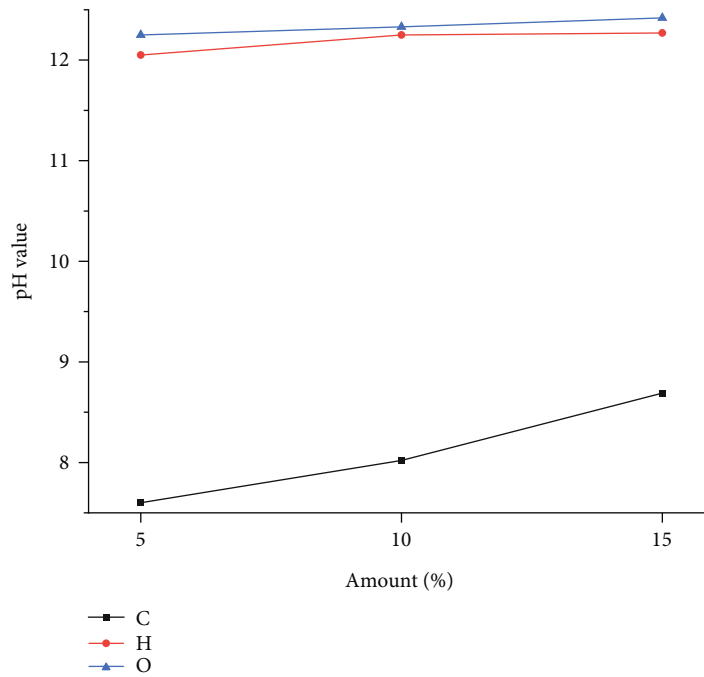
FIGURE 3: The unconfined compressive strength of each sample at 0.5% Zn²⁺ concentration.

of calcium hydroxide is similar to that of the samples with the same content of calcium oxide when the concentration of Zn²⁺ is increased from 0.1% to 0.5%. The strength of Z5H5 is higher than the compressive strength of Z5O5 as can be seen in Figure 2; however, with the increase in the amount of curing agent, the strength of the samples with the addition of calcium hydroxide is similar to that of the specimens with the same content of calcium oxide when the concentration of Zn²⁺ is increased from 0.1% to 0.5%. The strength of Z5H5 is higher than the compressive strength of Z5O5 as can be seen in Figure 3; however, with the increase in the amount of curing agent, the strength of the specimens with the addition of calcium hydroxide is less than the strength of the samples with the same content of added calcium oxide, although they are similar. This is probably due to the fact that after adding calcium hydroxide to the Zn²⁺-contaminated soil at a high concentration, there is enough Zn²⁺ and OH⁻ in the soil, and CaZn₂(OH)₆·2H₂O generated by OH⁻, Ca²⁺, and Zn²⁺ in the soil has an obvious effect on the strength [28]; Zn²⁺ has an inhibitory effect on the hydration reaction [29], and the effect is more pronounced at high concentration, thus making Z5O5 generate a small amount of C-S-H and C-A-H, so that the strength of Z5H5 is greater than that of Z5O5; as the content of calcium oxide increases, calcium oxide generates more C-S-H and C-A-H in the soil, so that its strength grows rapidly and thus exceeds

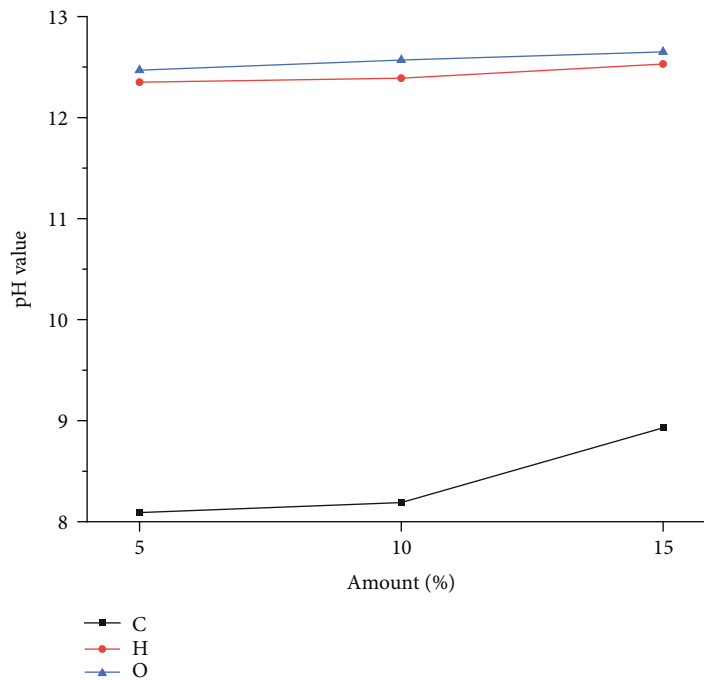
that of the samples with calcium hydroxide at the same content. The generation of C-S-H and C-A-H has a more obvious effect on the enhancement of the strength of the samples.

3.3. pH Test. The curing agents should not only provide strength for curing heavy metals in contaminated soils but also be friendly to the environment. Therefore, soil samples are tested for pH to detect the effect of the curing agent on the pH of the environment. Too high or too low pH can affect the survival of organisms or corrode buildings. Figure 4(a) shows the pH of each sample for a Zn²⁺ concentration of 0.1%, and Figure 4(b) shows the pH of each sample for a Zn²⁺ concentration of 0.5%. The pH of the samples with the addition of calcium carbonate was around 8. There was a large increase in pH with the increase in content, and the pH was close to 9 at 15% admixture. The addition of calcium carbonate resulted in a more moderate increase in pH [30]. The pH of calcium hydroxide and calcium oxide is closer, both around 12, and does not increase significantly with increasing content, and both calcium oxide is higher than calcium hydroxide.

After adding calcium hydroxide or calcium oxide, the pH is greater than 9, which causes damage to the environment again and forms acid-base pollution, and after adding calcium carbonate, the pH is less than 9, which is more friendly to the environment.



(a)



(b)

FIGURE 4: pH at different pollution concentrations. (a) pH value at 0.1% Zn²⁺ concentration. (b) pH value at 0.5% Zn²⁺ concentration.

3.4. XRD Analysis. Soil samples with a contamination concentration of 0.5% and a curing agent content of 10% were analyzed for XRD physical phase composition, as shown in Figure 5, in which all three different curing agents have an identical and more obvious diffraction peak of SiO₂. Adding CaCO₃, another obvious diffraction peak of

CaCO₃ shows. Adding Ca(OH)₂ can find Ca(OH)₂ and CaZn₂(OH)₆-2H₂O diffraction peaks. The diffraction peaks of the products CaCO₃, Ca(OH)₂, CaZn₂(OH)₆-2H₂O, C-S-H, and C-A-H can be found after the addition of CaO. It indicates that CaZn₂(OH)₆-2H₂O is formed after the addition of Ca(OH)₂; CaCO₃, Ca(OH)₂, CaZn₂(OH)₆-2H₂O, C-S-H,

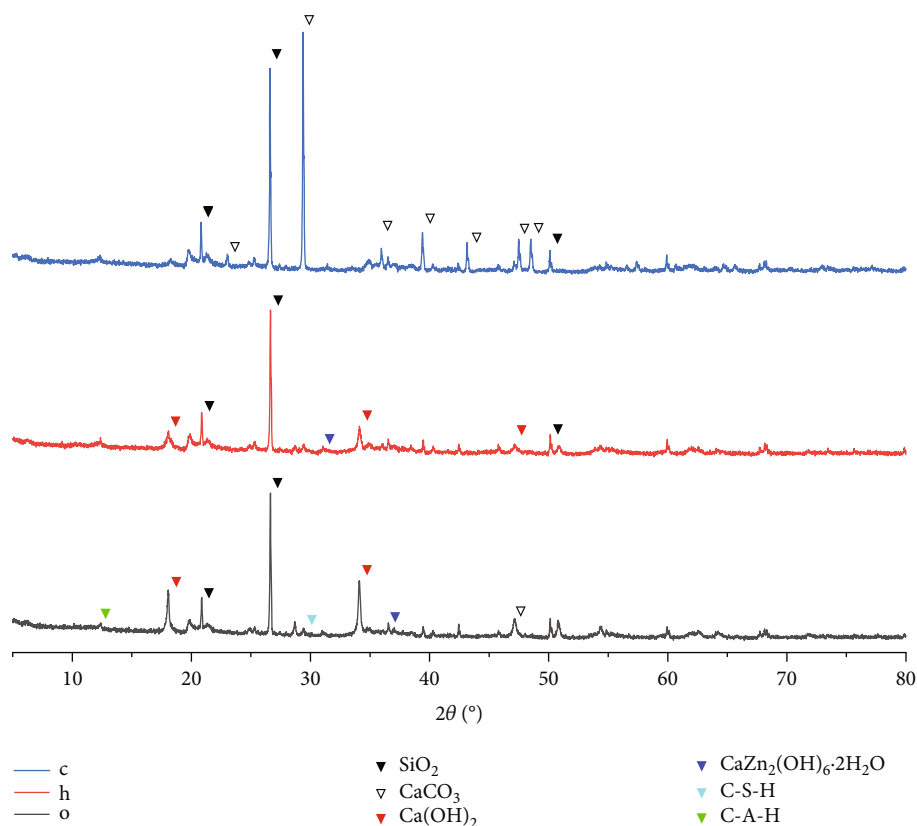


FIGURE 5: XRD patterns of zinc-contaminated red clay cured by different curing agents.

and C-A-H are formed after the addition of CaO. This reinforces the above inference that the production of these products increases the strength of the specimen.

4. Conclusions

Based on the results and discussions presented above, the conclusions are obtained as below:

- (1) After adding calcium oxide to Zn^{2+} -contaminated red clay, calcium oxide will react with SiO_2 , Al_2O_3 , and Fe_2O_3 in the red clay to produce C-S-H and C-A-H, which has a better effect on the strength of the soil, better for the low concentration of contaminated soil and weaker for the high concentration of contaminated soil
- (2) Calcium carbonate has a weakening effect on the red clay contaminated with zinc; the strength of the contaminated soil with calcium carbonate added is lower than that of the contaminated soil without calcium carbonate. The strength of Zn^{2+} -contaminated red clay with the addition of calcium hydroxide has a significant increase; the strength of contaminated soil with a high concentration of calcium hydroxide is higher. It is because there are enough zinc ions to react with hydroxide to form $CaZn_2(OH)_6 \cdot 2H_2O$
- (3) The pH regulation of calcium carbonate for zinc-contaminated soil is relatively mild, and pH is

around 8, which is a partial alkaline environment. And the pH increases a lot after mixing with calcium oxide and calcium hydroxide, around 12, which has a strong alkaline environment and causes secondary pollution to the environment

Data Availability

Data supporting the results of our study are in the article.

Conflicts of Interest

We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

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