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Experimental Study on Strength Characteristics of Zinc Contaminated Soil Solidified by Cement-Metakaolin-Limestone

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Abstract: In order to reveal the effect of curing agent ratio variable and curing age on zinc heavy metal contaminated soil solidified by cement-metakaolin-limestone (LC^3) ternary mixture, artificial contaminated soil and curing agent were used to simulate the solidification of contaminated soil. Through a series of laboratory tests, the change rules of unconfined compressive strength and electrical conductivity after curing were analyzed. It is found that with the increase of curing age, the stronger the compressive strength is, the lower the moisture content is. The compressive strength of solidified soil decreases with the increase of moisture content, showing an obvious correlation. Thus it can be seen that the coupling substitution of high territory and limestone for cement can obtain lower moisture content and higher strength and excellent mechanical properties at a relatively early age. The pH value of solidified soil increased to 9.68-10.78. Considering many factors, the solidification effect of zinc heavy metal is the best when the ratio of cement, metakaolin and limestone is LC^3-3.

Keywords: Solidification/stabilization, Zinc-contamination, Soils, LC^3, Strength characteristic.

1. Introduction

With the rapid development of economy, the content of heavy metals in industrial sites and other soils seriously exceeds the standard, and heavy metal pollution has strong toxicity, complexity, persistence, concealment and irreversibility[1, 2]. Zinc is an essential trace element in organisms, but its excessive accumulation in soil ecosystem will worsen soil physical, chemical and biological characteristics, lead to soil degradation, and reduce the quantity and quality of crops[2]. According to the National Environmental Protection (site pollution Assessment) Standard (1999), the recommended lower limit concentration of Zn in soil and groundwater (ecological survey level) is 200mg/kg. Therefore, it is very necessary to identify and implement cost-effective remediation technology to deal with heavy metal contaminated soil. Subsequently, the restored soil can be reused as sustainable engineering materials for infrastructure construction such as roads and buildings.

At present, the most mature and widely used method for remediation of heavy metal contaminated sites is solidification / stabilization (solidification/stabilization,S/S), which will fix heavy metal ions and prevent them from further diffusion to the surrounding environment through the physical and chemical reactions among heavy metals, soil and curing agents[3]. Commonly used curing agents include cement, lime and fly ash. However, high concentrations of heavy metals will seriously inhibit the hydration of cement-based materials, resulting in a decrease in the number of hydration products, resulting in a decrease in the strength of solidified contaminated soil and a surge in the dissolution of heavy metals[4]. At the same time, the global supply of auxiliary cementitious materials such as lime and fly ash is limited, which hinders them from meeting the growing market demand[5]. Therefore, efficient, low-cost and environmentally friendly materials are needed as curing agents for the remediation of heavy metal contaminated soils.

ScrivenerK et al.[6] proposed a cement-metakaolin-limestone ternary system (Limestone calcined clay cement referred to as LC^3). In order to reduce carbon dioxide emissions and energy consumption in the cement industry, LC^3 can be used as a curing agent to stabilize and repair heavy metal pollution, which can effectively promote the development of industrial pollution treatment and remediation technology in China. In this paper, using artificially prepared Zn contaminated soil, cement-metakaolin-limestone was applied to the solidification of Zn-contaminated soil. Through the tests of unconfined compressive strength (UCS), water content and soil pH value, the effects of curing agent ratio and curing age on the solidification of Zn-contaminated soil by cement-metakaolin-limestone (LC^3) ternary mixture were studied.

2. Materials and Methods

Materials. The test soil without pollutants is taken from the soil near Zigong City, Sichuan Province. the test soil is a typical low liquid limit clay. The optimum moisture content is 20.05%, the maximum dry density is 1.712g cm-3, the plastic limit is 18.1%, and the liquid limit is 29.55%. The curing agent materials are Portland cement, metakaolin and limestone, and the test water is deionized water. Because nitrate ion has little effect on cement hydration reaction. Therefore, heavy metal pollutants are made by adding zinc nitrate hexahydrate solution. Choose the method of artificial preparation of heavy metal contaminated soil[7].

Sample preparation. In this study, the amount of curing agent (the ratio of total mass of curing agent to dry soil) is 6%. According to the investigation of heavy metal pollution in typical industrial polluted sites, the concentration of heavy metals (the ratio of Zn mass to dry soil mass) reached 5000mg/kg. The moisture content of the sample is 25% (slightly larger than the optimal moisture content of uncontaminated soil). Samples of contaminated soil, Portland cement, metakaolin and limestone were prepared according to different components (Table 1). After preparation, the samples were cured under standard curing conditions (temperature 20 ±2 ℃, relative humidity above 95%) for 7 days and 28 days.

Testing methods. The unconfined compressive strength test is carried out with reference to "Highway Geotechnical Test Code JTGE40-2007". Take about 20g soil sample from the center of the sample, put it in a weighing bottle, bake it in an oven at 105 ℃ for 8 hours, dry until weighed, and calculate the moisture content of the sample. Then some residual soil was sieved by 2mm, and the pH test was carried out. 10.00 g of residual soil was sieved by 2mm and added with 10mL deionized water according to the proportion of 1:1. The pH value was measured after stirring evenly and standing for 1 h.

Table 1. Composition of curing agent with mass ratio

Materials	Portland cement	Metakaolin	Limestone	Group No.
LC^3-1	55%PC	30%MK	15%LS	$PC_{55\%}MK_{30\%}LS_{15\%}$
LC^3-2	45%PC	40%MK	15%LS	$PC_{45\%}MK_{40\%}LS_{15\%}$
LC^3-3	35%PC	50%MK	15%LS	$PC_{35\%}MK_{50\%}LS_{15\%}$
LC^3-4	45%PC	30%MK	25%LS	$PC_{45\%}MK_{30\%}LS_{25\%}$
LC^3-5	35%PC	30%MK	35%LS	$PC_{35\%}MK_{30\%}LS_{35\%}$

3. Results and Discussion

3.1. Moisture content of solidified soil

6% LC3 solidified samples with different components were selected for moisture content test. The formula for calculating moisture content ω:

$$\omega = \frac{M_w}{M_s} = \frac{m_1 - m_2}{m_1 - m_0} \times 100\% \qquad (1)$$

In the formula, ω is the moisture content of the sample (%), m_0 is the container mass (g), m_1 is the total mass of the sample and container before drying (g), and m_2 is the total mass of the sample and container after drying (g).

The results of moisture content ω of LC^3 cured samples with different components are shown in Figure 1. As expected, the moisture content decreased continuously with the extension of curing time, by 5.71-25.3%. This shows that with the increase of time, the hydration reaction of curing agent becomes more and more sufficient, and the free water is consumed. The content of metakaolin or limestone in the curing agent increases and the water content decreases, indicating that the surface area of metakaolin mineral is large, and the main component kaolinite has good water absorption capacity. At the same time, limestone itself has good mineral water absorption, which offsets the slow decline rate of water content caused by the lack of $Ca(OH)_2$ when Portland cement is reduced.

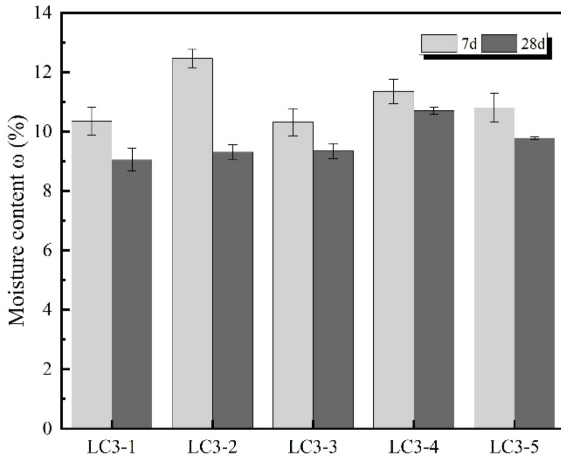

Figure 1. Effect of LC^3 solidification of Zn contaminated soil with different components on water content.

3.2. pH value of solidified soil

Figure 2 shows the pH of Zn-contaminated soil solidified by different components of LC^3 curing agent. It is found that the addition of curing agent can significantly increase the pH of Zn-contaminated soil, and the pH value of Zn-contaminated soil changes from 5.87 to 9.68-10.78, which makes Zn-contaminated soil change from acidic to weakly alkaline, and heavy metals can be adsorbed in the process of forming various insoluble metal hydroxides[8]. The reason may be that the basic substances in the curing agent (that is, CaO in the binder) are hydrated to form OH^- ions, so pH in the treatment system supports the formation of various hydration products with the increase of time, such as $Ca(OH)_2$, AFt $[Ca_6Al_2(SO_4)_3(OH)_{12}\cdot 26H_2O]$, C-S-H and $CaCO_3$. Also from the figure, we can see that the pH value of solidified soil increases with the increase of curing age, but the change range is small. With the increase of the content of metakaolin or limestone, the pH value decreases slightly, which may be due to the gradual consumption of alkaline substances by the volcanic ash (mainly SiO_2) in kaolin and soil[9]. The pH values of all samples meet the standard value of pH < 12.5 for hazardous wastes (GB5085.3-2007)[10].

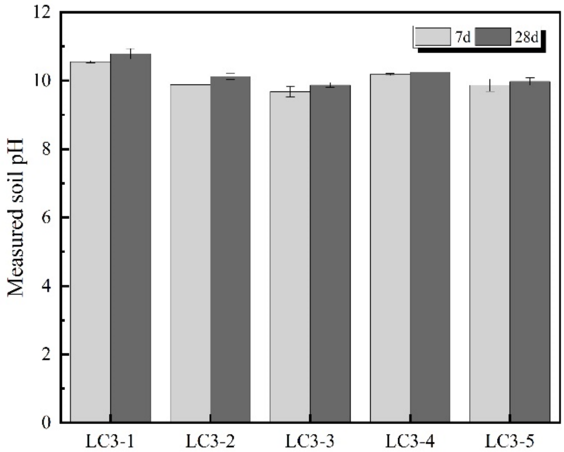

Figure 2. Effect of different components of LC3 on pH value of Zn contaminated soil

3.3. Unconfined compressive strength of solidified soil

The unconfined compressive strength of Zn-contaminated soil cured by different components of LC^3 curing agent is shown in Figure 3. It can be seen that the strength of solidified soil after 28 days of restoration is much higher than the design code limit 350 kPa required by USEPA[11], and exceeds the minimum unconfined compressive strength required by European countries such as the Netherlands, the United Kingdom and France, 1Mpa[12].

From Figure 3, it can be seen that the compressive strength of all samples increases with the curing age, with an increase of 5.53-23.8%. This is mainly due to the continuous hydration of the ternary mixture and the reaction of calcium carbonate in limestone and alumina / aluminate in metakaolin to form calcium aluminate AFm hydration products with a structure similar to ettringite, stabilizing ettringite, ensuring better space filling and exquisite pore structure[13], which can counteract part of the blocking effect of heavy metals, and then improve its strength.

Figure 3 shows that from LC^3-1, 2, 3 samples, with the increase of the proportion of metakaolin, the cement content decreases and the strength increases gradually at all ages. It may be due to the pozzolanic reaction and the increase of the formation of carbonate-AFm phase in the ternary binder, which will lead to the enhancement of strength. With the increase of the proportion of limestone, the strength also increases to a small extent, which may be due to the increase of limestone, accelerate the hydration reaction, form hydration products and fill pores. It is found that the strength of LC^3-3 at 7d and 28d is better than that of other ratios, the mechanical properties are better, and the curing effect may be better.

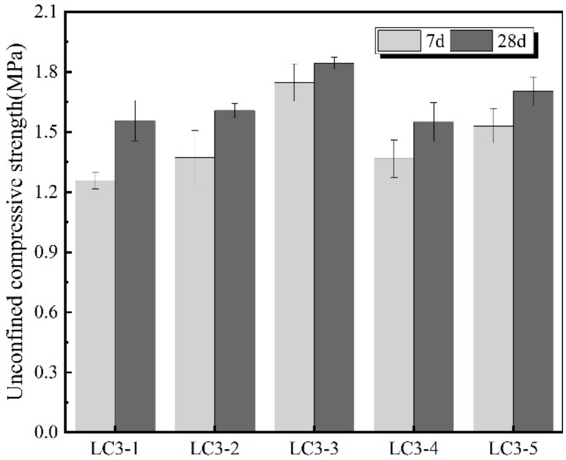

Figure 3. Effect of different components of LC3 solidified Zn contaminated soil on unconfined compressive strength

3.4. Relationship between unconfined compressive strength and moisture content and pH value

The relationship between moisture content ω and unconfined compressive strength (q_u)of solidified soil is shown in Figure 4. The unconfined compressive strength of solidified soil decreases with the increase of moisture content. This is because the moisture content is too high and the moisture in the hydration reaction is too high, which can not only reduce the concentration of reaction ions in the internal environment of the solidified soil, weaken the solidification reaction[14], but also increase the fluidity of the soil. it reduces the bonding performance of hydration products to soil, resulting in the increase of particle spacing or the excessive spacing of particle clusters, resulting in the decrease of the strength of solidified soil.

Figure 5 shows the change law between pH value and unconfined compressive strength (q_u) of solidified contaminated soil, it can be seen that the test strength decreases with the increase of pH value; it may be due to the amphoteric characteristics of Zn, with the change of acid-base environment of the system, part of the hydroxides of Zn can gradually dissolve in water and interact with OH- to form zincate ion, which leads to the decrease of Zn strength with the increase of pH value.

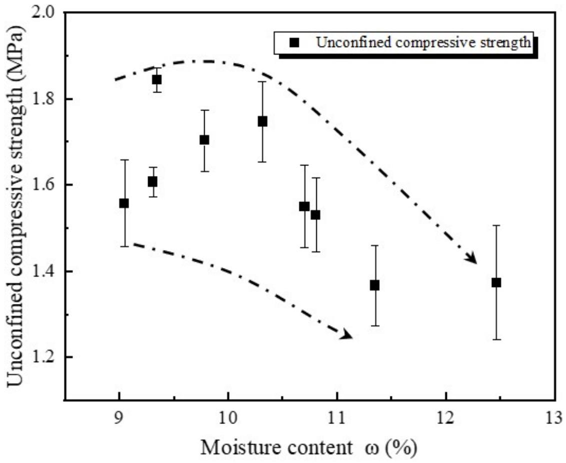

Figure 4. Fitting curve between unconfined compressive strength and moisture content of solidified Zn contaminated soil

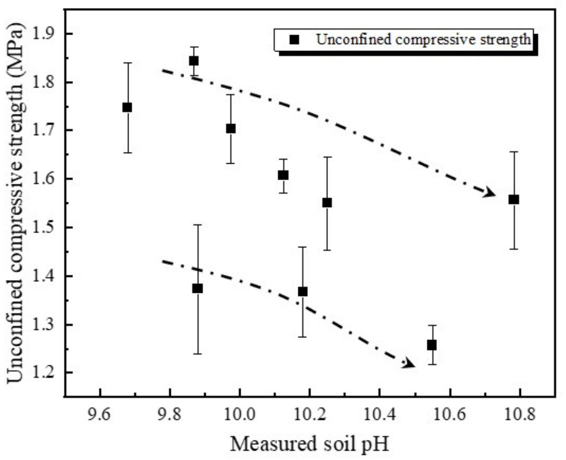

Figure 5. Fitting curve between unconfined compressive strength and measured soil pH of solidified Zn contaminated soil

4. Summary

The solidification effect of LC^3 composite cement material on Zn contaminated soil with different components was studied. Through unconfined compressive strength test, water content test and soil pH value test, the following conclusions were drawn.

(1) With the increase of time, the hydration reaction of curing agent becomes more and more sufficient, a large number of hydration products are formed, the matrix becomes denser and the water content decreases continuously.

(2) the pH value of soil mixed with curing agent changed from 5.87 to 9.68-10.78. The pH values of all samples meet the standard value of hazardous waste with pH less than 12.5.

(3) the unconfined compressive strength of solidified soil decreases with the increase of moisture content and pH value.

(4) the unconfined compressive strength of solidified contaminated soil increases with the increase of age. With the increase of the ratio of metakaolin to limestone, the strength increases gradually. The curing agent with the ratio of LC^3-3 has the highest strength.

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