Comparison of Compressibility Behaviour of Lime-Cement Stabilized Lateritic Clay Soil Contaminated by Heavy Metals

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Abstract. The type of stabilizer, heavy metal and soil can typically affect the strength and compressibility development in the treatment of contaminated soils. This is due to the difference in chemical reactions and interferences occurring among the stabilizers, metals, and clay minerals. A standard consolidation test using an oedometer was carried out to investigate the cement–lime induced changes in the compressibility of clay soils contaminated with either copper or zinc. In this study, laterite clay soil obtained from the hilly area at Universiti Teknologi Malaysia was used. Control samples of cement-and lime-stabilized soils without heavy metals were compared with the treated contaminated stabilized soil at various contaminants contents. Such percentage of contaminants means that the soil falls under industrially contaminated soil. To highlight the effect of the two heavy metals on the consolidation behavior of clay samples treated with cement and lime, a comparison has been made at a curing period of 7, 28, 100, and 200 days respectively. It was found that the interference of heavy metals during the process of hydration (cement and lime) contributed directly to the inhibition of the compression development of the tested soil samples. Therefore, it is crucial to identify the type of stabilizer, heavy metal, and soil involved to ensure successfully and remediate treatment.

1. Introduction

Nowadays, due to heavy industrialization, it is important to protect the environment from hazardous pollutants associated with waste generation and disposal. Several methods have been developed to reduce the potential release of toxic species into the environment or to transform the hazardous wastes into nontoxic substances. By using a chemical treatment, the stabilization and solidification processes can bind the hazardous waste compound to an established unsolvable System and catch the waste inside a compact cementitious medium, respectively [1]. The chemical reaction that arises from the interaction of waste components with a solidifying binder plays an important role that controls the quality of binder-based solidified waste products [2]. This method is commercially used in situ or ex. situ for treatment of contaminated soil because it is relatively cheap and results in well qualified long-term physical and chemical stability, which can increase the mechanical and structural characteristics [3]. Therefore, the
end products of such stabilized wastes can be reused for engineering fills, highway construction, pavement subgrade, backfill, base material, and so on.

The effectiveness of the stabilization and solidification process is dependent on the types of wastes and heavy metals. Portland cement using fly ash, lime, slag, and soluble silicates is among the solutions used in the chemical treatment of soil contaminated by heavy metal waste [4]. Ordinary Portland Cement (OPC) is the most commercially used solution as it is readily available, cost-effective, and also compatible with a variety of wastes [5-8]. However, several studies reported that the addition of heavy metals to the stabilized waste could retard the hydration process and, in some cases, inhibition of the cement hydration process occurred [9]. Cement hydration can also be delayed in the existence of zinc (Zn.), lead (Pb.), and copper (Cu.) when double salts are formed between these heavy metals and calcium ions that dissolve early from the cement [10 and 11]. Meanwhile, Tashiro and Oba, (1980) [12] reported that the heavy metal oxides of chromium (Cr), Cu, Zn, arsenic (As), and Pb influence the cement’s physical properties due to reactions with cement paste, which influence the cement paste desensitization and strengthening development during early stages of cement hydration.

Generally, the existence of heavy metals in the polluted cement-stabilized soil can retard the cement reactions. However, their effect on the reaction with lime has not yet been comprehensively studied. It should be noted that the lime in the form of (hydrated. Lime) is rather high, in Malaysia and has commonly been used in the treatment of tropical soils [13]. Therefore, in this study, lime was used as an alternative stabilizer in order to compare the efficiency and suitability of lime and cement as binders for stabilization/solidification (S/S) treatment of tropical laterite soils contaminated with heavy metals. The unconfined compressive strength of stabilized soil at different curing period intervals was determined to investigate the strength development for both cement and lime in contaminated stabilized soils. In addition, the one-dimensional consolidation (odometer) test was conducted to examine the compression index parameter for contaminated samples.

2. Materials and specimen preparation

The laterite residual soil used in this study was a reddish clay soil with a large quantity of iron oxides usually establish in humid areas. Table 1 shows (the chemical and physical properties) of the untreated soil. Hydrated lime was utilized in this research is It works well with clayey soils, particularly those with a reasonable to high plasticity index (PI > 15). In addition, OPC has been used as the second stabilizer in this work. Both substances were in powder form, and they were obtained from a factory in Pasir Gudang, Johor. The chemical compositions of the hydrated lime and Portland cement are shown in Table 2.
Table 1. The physical properties and chemical composition of laterite.

| Engineering and physical properties (laterite) | Values | Chemical composition oxides Values (%) |
|-----------------------------------------------|--------|----------------------------------------|
| CEC (meq/100 g)                               | 22.7   | SiO₂                                  | 21.55 |
| pH (L/S = 2.5)                                | 4.86   | Al₂O₃                                 | 24.31 |
| Specific gravity                              | 2.75   | Fe₂O₃                                 | 29.4  |
| Liquid limit, LL (%)                          | 72.8   | Na₂O                                  | 0.07  |
| Plastic limit, PL (%)                         | 35.6   | K₂O                                   | 0.11  |
| Plasticity index, PI (%)                      | 37.2   | P₃O₅                                  | 16.71 |
| BS classification                             | MH     | SO₃                                   | 3.98  |
| ICL (%) 2.00 soluble                          | 5      | CO₂                                   | 3.65  |
| Maximum dry density (Mg/m³)                   | 1.33   |                                       |       |
| Optimum moisture content (%)                  | 34     |                                       |       |
| Unconfined compressive strength (kPa)          | 288    |                                       |       |
| Cc                                            | 0.24   |                                       |       |

Note: ICL: Initial Consumption of Lime, Cc: Compression Index

Table 2. Chemical compositions of the hydrated lime and Portland cement.

| Hydrated lime | Chemical composition       | Values (%) | Chemical composition       | Values (%) |
|---------------|---------------------------|------------|---------------------------|------------|
| Calcium oxide, CaO | 74.23                    | Calcium oxide, CaO | 64.73                   |
| Phosphorus oxide, P₂O₅ | 0.08                     | Potassium oxide (K₂O) | 0.57                   |
| Magnesium oxide, MgO | 0.74                     | Magnesium oxide, MgO | 1.1                    |
| Calcium sulfate, CaSO₄ | 0.12                     | Sulfur trioxide (SO₃) | 2.01                   |
| Ferric oxide, Fe₂O₃ | 0.17                     | Ferric oxide, Fe₂O₃ | 3.1                    |
| Aluminium oxide, Al₂O₃ | 0.11                     | Aluminium oxide, Al₂O₃ | 5.2                    |
| Silica, SiO₂ | 0.14                     | Silica, SiO₂ | 21.1                    |
| Loss on ignition, LOI | 24.35                    | Loss on ignition, LOI | 0.8                     |
| Insoluble residue |                          | Insoluble residue | 0.1                     |

The components that were added to reproduce inorganic metal contamination were copper nitrate trihydrate (Cu (NO₃)₂ * 3 H₂O) to provide the copper (II) ions (Cu²+) and zinc nitrate tetrahydrate (Zn (NO₃)₂ * 4 H₂O) to provide zinc (II) ions (Zn+2). Both of them were from Merck (KGaA), Germany. The two substances were selected due to their occurrence at many polluted situates; their chemical and physical properties are as listed below in Table 3.
Table 3. The chemical and physical properties of the selected reagents

| Metal                        | Chemical and physical properties | Metal                        | Chemical and physical properties |
|------------------------------|----------------------------------|------------------------------|----------------------------------|
| Copper dinitrate trihydrate  | Cu(NO₃)₂ * 3 H₂O                 | Zinc nitrate tetrahydrate    | Zn(NO₃)₂ * 4 H₂O                 |
| Solubility                   | 2670 g/l (20 °C)                 | Solubility                   | (20 °C) soluble                  |
| Melting point                | 114 °C                           | Melting point                | 45 °C                            |
| Molar mass                   | 241.60 g/mol                     | Molar mass                   | 261.44 g/mol                     |
| Density                      | 2.05 g/cm³ (20 °C)               | Density                      | 1.0 g/m³                         |
| Bulk density                 | 1050 kg/m³                       | pH value                     | 5 (50 g/l, H₂O, 20 °C)           |
| pH value                     | 3–4 (50 g/l, H₂O, 20 °C)         |                              |                                  |

Preceding studies accompanied on lateritic soil have shown that oven drying can significantly modify the plasticity and compaction properties of the soil [14]. Consequently, in this study, all the soil samples were prepared from air-dried soil cracked into lesser sizes and sieved through a 2 mm sieve. The sieving was complete to confirm that the soil was of uniform grade.

The concentration of stabilizing ions has a great influence on the short and long term reactions respectively during the initial modification and the cementation of the treated samples. Based on [5 and 16], concentration of lime between 2 and 3% of the soil’s dry weight is adequate for amendment purposes, and for stabilization purposes, the concentration typically ranges from 5 to 10%. The latter range is more prominent for pozzolanic reactions. Kassim and Chern, (2004) [13] discovered that, for typical Malaysian cohesive soils, the optimum lime content for stabilization should be around 3 to 6%. Due to the studies on improving the contaminated soils are relatively less, thus, the suitable percentages of contamination-stabilizers were based on the percentage of pozzolanic reactions. For comparison purposes, two equivalent contents of stabilizers were used (5 and 10%) for both lime and cement in this study.

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The essential quantity of water or optimum water content (OWC) depends on nature of the soil and stabilizer content In BS.1377: Part 4: 1990 (Clause 3.3.4.1) [17 and 18], five sequence of standard Proctor compaction tests are mentioned to ascertain OWC for several lime and cement mixtures. In this study, the mix proportions (percentage lime, percentage cement) were (0, 0), (5, 0), (10, 0) and (0, 5), (0, 10) as percentages by weight of dry soil. Depending on these percentages the labels of samples for lime and cement treated laterite clay soil are (LCLT5, LCLT10, LCCT5, and LCCT10) respectively. Should be noted that (LC) mentions to lateritic clay, and (LT and CT) mention to lime and cement treated one after another. The optimum moisture contents were adopted in accordance with these mix proportions as the calculated water content of contaminated soil samples. Soil samples artificially polluted with heavy metals were equipped by mixing zinc and copper nitrate as the sources of contaminants into the natural laterite clay. Three percentages of heavy metals were nominated, namely 0.1, 5. and 10% (dry soil weight basis). These different percentages were based respectively on the level of pollutants commonly found in naturally contaminated soil, industrially contaminated soil, and highly contaminated soil. Control samples (cement- and lime-treated soils without heavy metals) were as well set for evaluation purposes.

To taste the contaminated the soil, all the laboratory instruments and the sampling device were previously immersed in a 5% nitric acid solution, followed by deionized water for a day before sampling in order to remove traces of metals. Through the preparation of the sample, lime and cement were first individually mixed with the dry medium to certify a homogeneous distribution of the two stabilizers in the soil. Then, de-ionized water mixed with a pre-set quantity of copper nitrate or zinc nitrate was added to the mixture. Should be noted, only pure de-ionized water was used for the control samples. Based on laboratory trials, an additional quantity of water (2% of OWC) was added to all mix designs to take evaporation losses into account. After the mixing process, the soils were compacted in 38 mm × 76 mm
cylindrical moulds. The preparation process was concluded by making sure that the designed (O.W.C) and maximum dry Density according to Clause (4.1.5 of BS 1924: Part 2: 1990b) were achieved.

After that, the specimen was cured for two hours, removed from the mold, inserted into a thin-walled P.V.C tube, wrapped in a thin plastic lid and sealed with a tight rubber lid. Then, samples were kept to cure in a temperature controlled room (27 ± 2 °C) till the experiments were performed after 7, 14, 28, 100, and 200 days.

3. RESULTS AND DISCUSSION

Oedometer tests were carried out to evaluate the compressibility characteristics of cement- and lime-based lateritic clay soils contaminated with either 5% copper or 5% zinc. Such a contaminant percentage (5%) is commonly indicative of industrially contaminated soil. The curing time was considered to estimate the effect of heavy metals on the soil compression index. Here, a higher content of both stabilizers (10%) was selected to assess the effect of curing time on contaminated soil compressibility. To underscore the effect of both heavy metals on the consolidation behaviour of clay samples treated with cement, comparisons were made as shown in Figure 1(a–d) at curing intervals of 7, 28, 100, and 200 days, respectively. From the void ratio–axial pressure relationship of uncontaminated samples treated with cement (controls) and contaminated treated samples at 7 days, it is clear that the former group's compression began at a lower void ratio than that of the corresponding contaminated samples. However, the difference in void ratio between contaminated and uncontaminated samples decreased with curing time, as shown in Figure 1(b–d). This was most likely due to cation exchange reactions, where an increase in both flocculation and aggregation caused a chemically induced 'pre-consolidation' effect, which increased the vertical effective yield stress and reduced the compressibility characteristics [19 and 20].

Figure 2 reveals variations in the compression index of cement-treated clay soil with and without heavy metals over time. This is in agreement with [21], who reported that the compressibility of treated homogeneous specimens was much less than that of untreated clay. This implies that cement-treated clay undergoes structuration due to the effect of cementation up to its apparent yield stress level, after which restructuration occurs. As for heavy metals’ effects on the compressibility of treated contaminated clay samples, it was observed that their presence increased the compressibility of contaminated clay soils and that this increase was dependent on the heavy metal type. Zinc had a greater effect on the increase of compressibility than did copper, most likely due to its chemical reactions with calcium hydroxide and clay minerals. A gradual reduction in Cc of contaminated clay over time was most likely the result of soil hardening, mainly due to an increase in the amount of pozzolanic reaction compounds over time, which, in turn, reduced heavy metal inhibition.
Figure 1. Relationship of void ratio with axial pressure of uncontaminated and copper/zinc-contaminated cement-stabilized laterite clay at different curing times: (a) 7 days, (b) 28 days, (c) 100 days, and (d) 200 days.

Figure 3 illustrates the consolidation behaviour for lime-stabilized lateritic clay soil in comparison with heavy-metals-contaminated lateritic clay treated by the same stabilizer. The comparison has been applied with respect to changes in the curing time, as shown in Figure 3(a–d), at 7, 28, 100, and 200 days respectively. Figure 3(a) shows that the consolidation curve of lime-treated soil started at a higher void ratio than that of cement-treated clay samples, as observed in Figure 1(a), which means that cement is more suitable for improving the compressibility characteristics of lateritic clay soil.
Figure 2. The variation of the compression index of uncontaminated and copper/zinc-contaminated cement-stabilized laterite clay at different curing times.

Figure 4 depicts the evaluation using the compression index of lime-treated clay to estimate the effect of heavy metals on lime stabilization. As shown in this figure, it is obvious that the effect of heavy metals on the compressibility of contaminated lime-treated clay soil is different from the effect on cement-treated clay soil. Irrespective of the type of heavy metal, the compressibility of lime-treated clay decreased after both 7- and 28-day curing periods. The compressibility of uncontaminated treated clay and contaminated treated clay increased after 100 days due to an increase in the percentage void ratio resulting from the agglomeration and rearrangement of soil particles over time. However, a reduction in the compression index occurred at 200 days of curing time. This was attributed to the production of pozzolanic reactions, which formed a hardened skeleton matrix that enclosed the unaltered soil particles and led to strengthening of the soil and reduction of its compressibility. As such, it can be concluded that although heavy metals have less retarding effect on lime with regard to the compressibility of soil, cement is preferable for improving the compressibility of the contaminated soil.
Figure 3. Relationship of the void ratio with axial pressure of uncontaminated and copper/zinc-contaminated lime-stabilized laterite clay at different curing times: (a) 7 days (b) 28 days, (c) 100 days, and (d) 200 days.
CONCLUSIONS

On the basis of laboratory examinations, the following conclusions can be drawn:

- The results from the one-dimensional oedometer test showed that cement is more active than lime in reducing the compressibility of contaminated soil.
- Zinc has more effect on the increase of compressibility than copper due to its chemical reaction with calcium hydroxide and clay minerals. Moreover, zinc metal has an amphoteric nature which exhibit maximum solubility and then may affect the compressibility properties.
- Irrespective of the type of heavy metal, the compressibility of lime treated laterite clay has decreased after both 7 and 28 curing periods. The compressibility of uncontaminated treated clay and contaminated treated clay increased up to 100 days. The increasing in the compressibility may attribute the mechanism interparticulate forces in the soil particles which are occurred in the presence of non-expanding mineral such us kaolinitic mineral.

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