Mine Water and the Environment (2020) 39:103–111
https://doi.org/10.1007/s10230-020-00662-1

TECHNICAL ARTICLE

Effect of $CuSO_4$ on the Hydromechanical Behavior of Compacted Tailings

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Received: 15 May 2019 / Accepted: 22 January 2020 / Published online: 1 February 2020
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Abstract

Laboratory tests, including compressibility, permeability, and microstructure tests, were conducted on tailings samples using custom-designed test apparatus to investigate the effect of metal contamination (Cu^{2+}) on the hydromechanical behavior of compacted tailings. Infiltrating samples with various dry densities with distilled water or $CuSO_4$ solution at various concentrations showed that the void ratio of compacted tailings decreased with increased dry density. An increase in the metal contaminant concentrations from 0 to 0.1 mol/L increased the compression coefficient of the tailings from 0.14 to 0.84 MPa^{-1} under a vertical load of 0.01 to 2.0 MPa, while the yield stress of the tailings decreased from 204.3 to 98.7 kPa, respectively. The linear relationship between permeability coefficient (k) and void ratio (e) is described by $k = -6.48 + 17.17e$. Microstructure test results showed that the diffusion double layer thinned, and the surface potential decreased, indicating that the contaminant of Cu^{2+} enhanced the compressibility and permeability of the tailings. The microstructure test results also showed that the amount of fine-grained soil in the copper tailings was significantly less after the hydromechanical test. Therefore, the permeability and compressibility of copper tailings increased. The experimental results are in good agreement with the estimated results.

Keywords Copper tailings · Metal contaminant · Compressibility · Permeability · Fine-grained soil

Introduction

The demand for products of extractive industries is continuously growing across the globe. With the development of the mining industry, the disposal of industrial waste such as tailings is increasingly becoming a source of environmental pollution (Coulibaly et al. 2017; He et al. 2019a, b; Sharma and Busaidi 2001; Thomas and Gupta 2013; Wang et al. 2018). A tailings dam failure can pose potential hazards to the local ecology and human health (Byrne et al. 2018; Ferguson et al. 2009; Glotov et al. 2018; Kemper and Sommer 2002; Kossoff et al. 2014; Ngole-jeme and Fantke 2017; Queiroz et al. 2018; Villavicencio et al. 2013). Therefore, increased attention is being paid to the safety and environmental aspects of tailings ponds.

The geotechnical characteristics of tailings differ from those of conventional soil materials. Tailings have high angularity, uniform grains, poor gradation, complex composition, and metal content (Adajar and Miller 2018; Wilson et al. 2017; Zandarín et al. 2009; Zhang et al. 2015). The permeability and compressibility of tailings are directly related to their mineral composition, clay content, pore structure, stress state, deformation, self-weight, and pollutant contents of soils (Benjamin et al. 2005; Harehdasht et al. 2019; Salgueiro et al. 2008; Wen et al. 2018; Zhang et al. 2017). Many tailings pond failures are due to the influence of permeability and compressibility on tailings (Naeini and

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s10230-020-00662-1) contains supplementary material, which is available to authorized users.

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Akhtarpour 2018). Therefore, in the complex physical and chemical environment of a tailings pond, it is essential to study the effects of metal contaminants on the hydromechanical behavior of tailings.

The hydromechanical behaviors of compacted tailings have been studied under various conditions. Aubertin et al. (1996) showed that the permeability of tailings derived from a hard rock mine ranged from 10^{-5} to 10^{-4} cm/s, and was affected by the void ratio and grain size. The engineering characteristics of copper tailings were studied by Shamsai et al. (2007), who established a linear relationship between the permeability coefficient and void ratio. The mechanical properties of sulfur-bearing tailings were studied by Liu et al. (2013). The authors found that the tailings with higher sulfur content (sulfide and sulfate) had stronger compressibility, lower friction angle, and lower shear strength. Thomas and Gupta (2013) found that a grain composition range of 30% clay with 70% copper tailings to 70% clay with 30% copper tailings was the best combination to obtain good soil stabilization. Wickland et al. (2010) found that the permeability of a mixture of 5:1 waste rock to tailings was similar to that of the tailings, whereas the compressibility was significantly reduced. Wu et al. (2017a) found that tailings with various grain compositions had different permeability and compressibility characteristics. Moreover, based on an analysis of the critical clay content in tailings, Wu et al. (2017b) concluded that the strength of mixed tailings decreased gradually with an increase in the clay content. Adajar and Miller (2018) proposed that the hydro-compression settlement increases with an increase in pressure, up to the pre-consolidation pressure (or yield stress), after which it decreases, with pressures beyond the pre-consolidation pressure. A sample with low dry density below its optimum exhibits greater hydro-compression, which indicates that tailings have a degree of collapsibility.

The stress state and environmental conditions also affect the properties of tailings. Chu et al. (2016) studied the permeability characteristics of phosphogypsum tailings and found that when the actual hydraulic gradient of a tailings dam was greater than the critical hydraulic gradient, the tailings dam suffered a permeability deformation or an instability failure. Alhomair et al. (2016) found that adding fly ash to clay tailings increased its permeability, which they attributed to the amount of clay minerals. Chen et al. (2018) found that the permeability coefficient of tailings was positively related to compressive pressure and directly determined by the dry density of soil and the concentration of pore water. Based on the soil–water characteristic curve (SWCC), Zhang et al. (2018) found that changes in the void ratio and the degree of saturation of oil sands tailings were key factors that influenced permeability, and the fitting of the SWCC affected the quality of the estimated permeability function. He et al. (2019c) had similar results with compacted bentonite.

Chemical solutions can influence the hydromechanical behavior of tailings. Luan and Tang (1993a, b) studied the sorption of Cu, Cd, and other metal ions by tailings and showed that the tailings have a strong sorption capacity for dissolved metal ions. Wiertz and Marinkovic (2005) found that hydraulic diffusion largely controls metal contaminant transport in tailings pond. Elza et al. (2006) concluded that differential hydrology induces variable metal speciation in Pb–Zn mine tailings. Wu et al. (2008) studied the influence of Fe ions on the permeability of tailings. The authors found that metal ions are often transported along with groundwater seepage, but that during the migration of these metal ions, a series of redox reactions occur. These generate sediments in the tailings, which changes the seepage velocity and increases the possibility of seepage failure of the dam. Jin et al. (2017) studied the effect of an acid-based solution on the mechanical properties of sulfur-bearing tailings, and found that the combination of dissolution reactions and chemical precipitation increased the porosity of the tailings, which gradually increased the compressibility and permeability.

Few studies have comprehensively focused on the mechanism of the effects of metal contaminants on the hydromechanical behavior of compacted tailings. Therefore, we studied the effect of a metal contaminant (Cu^{2+}) on the compressibility and permeability of tailings. Using tailings from the Cheng-men Mountain copper mine in Jiangxi Province, China, we measured the compressibility and permeability of compacted tailings samples with different dry densities. Various concentrations of Cu^{2+} solution were infiltrated into the tailings for saturation and permeability testing, and the effect of the Cu^{2+} on the hydromechanical mechanism was studied relative to the microstructure and mineral composition of the tailings.

Materials and Methodology

Materials

The tailings were sampled from the tailings pond of the Cheng-men Mountain copper mine in Jiujiang City, Jiangxi Province, China (Fig. 1). The mining area is located in the lower reaches of the Yangtze River, bordering Ba-li Lake and passing through the Rui-chang River in the west, which is a tributary of the Yangtze River. At present, three tailings ponds have been built on the east side of the mine, about 1.5 km away from the linear distance of the concentrator: the Xiong-jia-yao, Feng-zhao-gou, and Liu-jia-gou tailings ponds. The service period of the first two tailing ponds has expired, and they are completely closed. The Liu-jia-gou tailings pond is in use, with a total storage capacity of 1640×10^4 m^3.

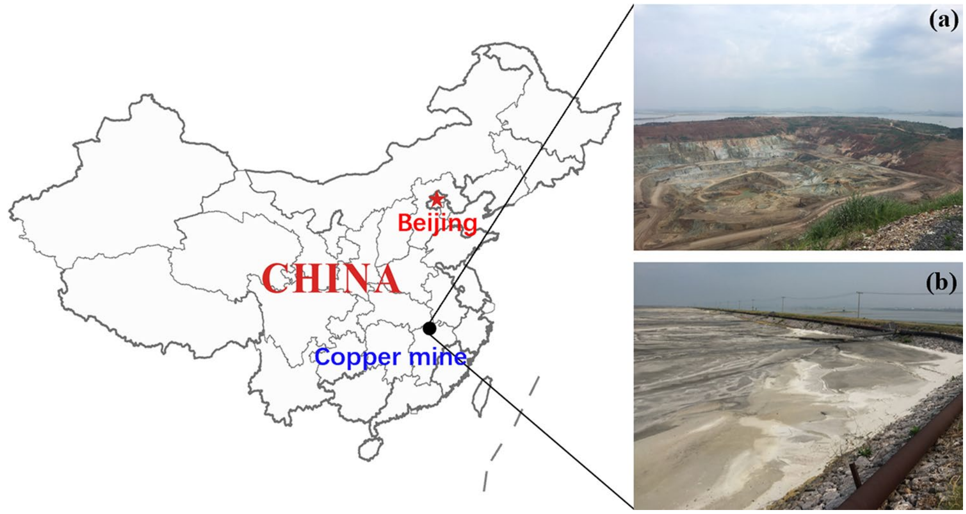

Fig. 1 Location of the study area. **a** copper mine; **b** copper tailings pond

Table 1 Basic physical and mechanical properties of tailings

Property	Value
Natural water content (%)	16.44
Dry density (Mg/m^3)	1.28
Specific gravity/G_s	2.77
Natural void ratio	0.538

The physical and mechanical properties of the tailings were tested in accordance with the ASTM E1856-97(2002) standard. The results are presented in Table 1. The chemical solutions used in this study were of analytical grade with a purity of 99%. The $CuSO_4$ solution was prepared using its sulfate. The conductivity of the distilled water was EC ≤ 4 μS/cm.

Sample Preparation

According to the required dry density, tailings of the corresponding weight were poured into a ring of the sample. Then, the sample was compacted to the designed height using a numerically controlled mechanical press with a vertical loading speed of 0.2 mm/min. To prevent the sample from rebounding, a constant height was maintained for 30 min, producing a standard sample with a 50 mm height and a 50 mm diameter. The error of the dry density of the compacted sample was ± 0.1 g/cm^3. Permeability and compressibility tests were performed on compacted reconstituted samples of the tailings (Supplemental Figure S-1).

Experimental Investigations

Permeability Test

We conducted a permeability constant head test using a custom-designed permeability cell (Fig. 2). According to

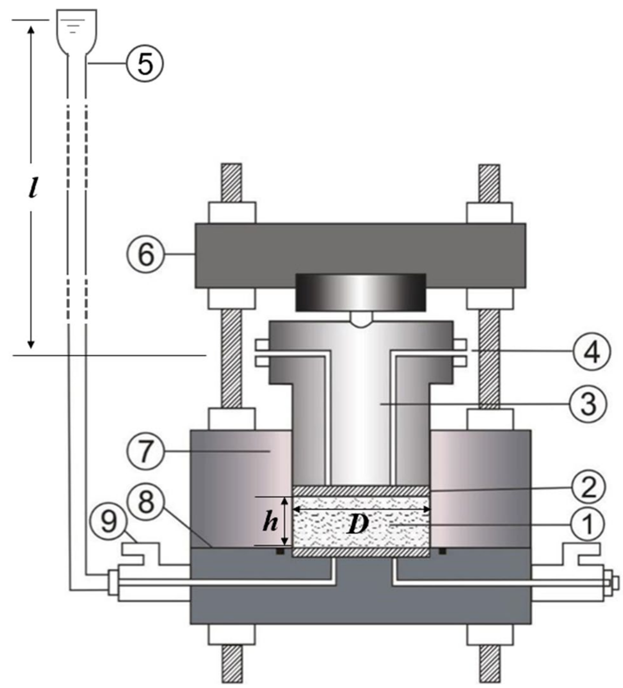

Fig. 2 Cell for the permeability test (① sample ② porous plate ③ piston ④ drainage valve ⑤ waterhead ⑥ top cover ⑦ cell ⑧ pedestal ⑨ injection valve)

Darcy's law, the expression for the permeability coefficient is

$$k = \frac{Q}{A \cdot J} = \frac{4hV}{\pi l t D^2}, \tag{1}$$

where k is the permeability coefficient (m/s), Q is the volume rate of flow (m^3/s), A is the cross-sectional area of the sample (m^2), J is the hydraulic gradient, V is the seepage volume (m^3) within the seepage time t (s), h is the seepage path length (m), l is the head height (m), and D is the sample diameter (m).

After compaction, the as-compacted tailings sample was introduced into the testing apparatus, as shown in Fig. 2. Solutions of various concentrations were infiltrated through the porous plate at the bottom of the sample. The evolution of the volume rate of flow was recorded. When the solution flooded out from the top of the instrument with a constant flow (④ in Fig. 2), equilibrium was assumed and the infiltration test was completed. This procedure was repeated on samples with different dry densities infiltrated with distilled water (Supplemental Table S-1). Three concentrations (0.01, 0.05, and 0.1 mol/L) were used to study the influence of solution concentrations on the permeability of the compacted tailings with a dry density of 1.6 Mg/m^3.

Compressibility Test

Compressibility tests were conducted on tailings samples with a custom-designed high-pressure oedometer (Supplemental Figure S-2). The high-pressure oedometer mainly comprises a loading frame, an oedometer cell, and a solution circulation system. The setup can provide a range of vertical stresses from 0.01 to 5.0 MPa. Details of the high-pressure oedometer can be found in He et al. (2016).

To study the influence of the Cu^{2+} on the compressibility of saturated tailings, the consolidation curves of tailings under various concentrations of the $CuSO_4$ solution were obtained using the rapid consolidation method. During the test, the sample and porous plate were installed onto the oedometer cell. A vertical stress of 0.01 MPa was applied for good contact between the sample and the test apparatus. It should be noted that this applied vertical load was much less than the compaction stresses the sample experienced. Thus, the sample remained in an over-consolidated state. Vertical deformations of the sample were recorded using a strain gauge with a precision of 0.01 mm for a total run of 25.0 mm during the test. The void ratio during the loading process was calculated using the initial void ratio of the sample and the measured vertical deformation. The details on sample dry density and solutions used in the compression test are shown in Supplemental Table S-2.

According to Terzaghi's one-dimensional consolidation theory, the initial void ratio of the tailings can be calculated by:

$$e_0 = \frac{G_s \cdot \rho_w}{\rho_d} - 1 \tag{2}$$

where G_s is the specific gravity of tailings in Table 1, ρ_w is the dry density of water (Mg/m^3), and ρ_d is the dry density of tailings (Mg/m^3). With the measured vertical deformation, the void ratio during the loading process can be calculated using Eq. (3):

$$e = e_0 - \frac{s}{H_0}\left(1 + e_0\right) \tag{3}$$

where s is the settlement at a certain time, H_0 is the initial height of the sample, and e_0 is the initial void ratio.

All tests were performed at a controlled ambient temperature of 20 ± 1 °C.

Microstructure Tests

XRD Test Tailings samples were pressed in stainless steel sample holders for the XRD analysis. A 12-kW rotating anode X-ray diffraction meter, D8 FOCUS (Germany, Bruker), operating at 30 mA and 40 kV, was employed. The XRD analysis was performed with CuKx radiation (λ = 0.15418 nm). The measurement range was from 5° to 85°, with a scanning rate of 8° (2θ) min^{-1}. The JCPDS PDF database was used for the phase identification.

Grain Size Distribution Test The grain size distribution of the tailings was investigated by an automatic laser particle size analyzer (Rise-2002), operating at 50 Hz and 200 W. A He–Ne gas laser source (λ = 0.6328 μm) was used. The measuring range was 2–2000 μm. The maximum detection angle was 105°.

Results and Discussion

Characterization

The XRD pattern for the copper tailings powder (Supplemental Figure S-3) indicates that the mineralogical composition of the tailings is quartz, orthoclase, sanidine, silicon oxide, and kaolinite. The grain size distribution of tailings is shown in Fig. 3, and the tailings group allocation is presented in Table 2.

As shown in Fig. 3 and Table 2, the samples were mainly composed of silt and fine sandy tailings. Furthermore, fine-grained soil comprised 83.8% of the total. The uniformity coefficient was less than 5, and the curvature coefficient was greater than 1.

Compressibility of the Compacted Tailings

Compression curves of the saturated tailings samples with various concentrations of the $CuSO_4$ solution are shown in Fig. 4, which clearly indicates that the tailings sample in distilled water exhibited less compressive deformation. Meanwhile, the tailings sample in the $CuSO_4$ solution had a larger compressive deformation, and the maximum

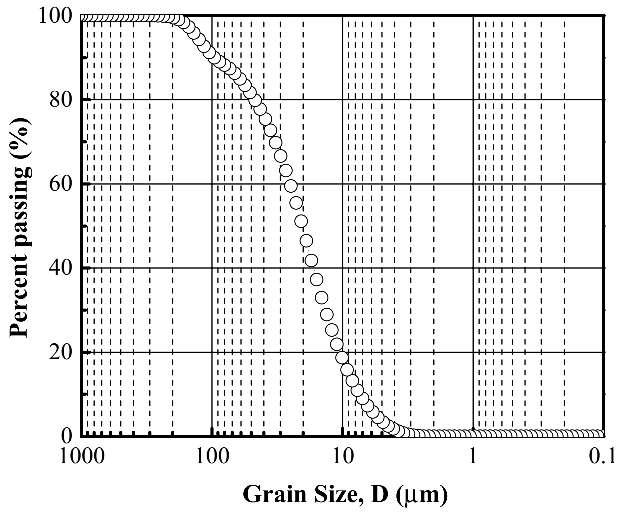

Fig. 3 Grain size distribution of tailings

Table 2 Tailings group allocation

Name	Grain size/μm	Percent passing/%
Medium tailings	250–850	0.98
Fine tailings	75–250	15.24
Fine silt tailings	5–75	79.75
Clay tailings	<5	4.03

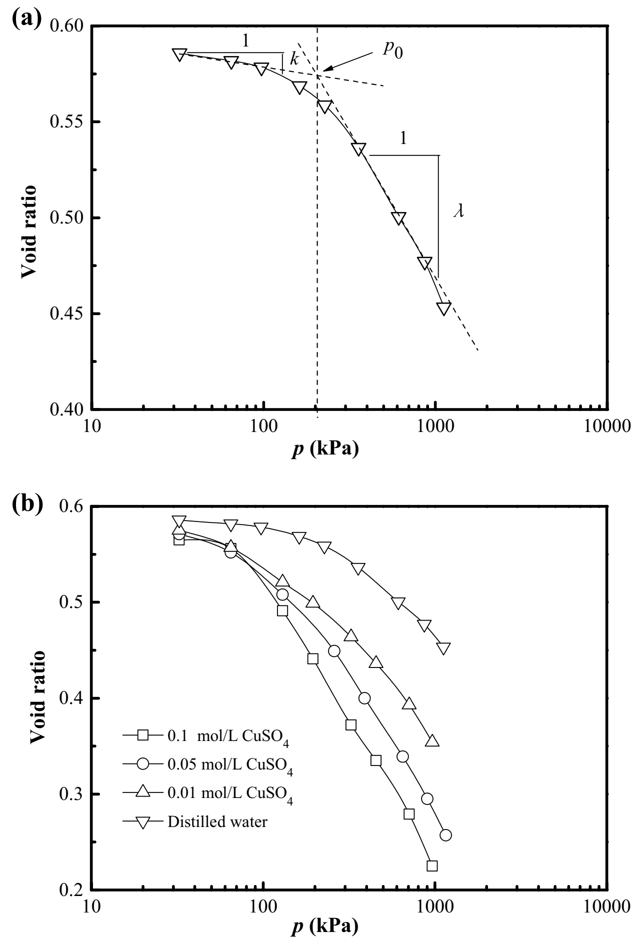

Fig. 4 Compression curves of compacted tailings **a** typical compression curve; **b** with varying concentrations of $CuSO_4$

deformation was positively correlated with the solution concentration. During the loading process, particle movement near the interface of the silt tailings was more evident, and the clay entered the pores of the silty sand tailings, compacting the grain structure of the tailings, and continuously reducing its compressibility (Zhang et al. 2015).

Compression test results are drawn in the *e*-log*p* plane (Fig. 4a). The entire compression curve is divided into an elastic deformation stage and a plastic deformation stage. The slope of the elastic deformation stage is defined as the elastic compressibility coefficient (k); the slope of the plastic deformation stage is the plastic compressibility coefficient (λ). The abscissa of the intersection of the two fitting lines of the elastic deformation stage and the plastic deformation stage depict the yield stress (p_0). As shown in Fig. 4a, considering the tailings sample in distilled water as an example, the yield stress of the tailings was calculated according to the stress–strain curve, and the compressibility of tailings was characterized. The yield stresses of compacted tailings with different concentrations of $CuSO_4$ are shown in Fig. 5. Figure 5 shows that the yield stress of tailings decreases significantly from 204.3 to 98.7 kPa with an increase in the concentration of $CuSO_4$.

The compression indexes of tailings samples treated with $CuSO_4$ solution and distilled water are compared (Fig. 6). The compression index (C_c^*) is defined as the ratio of $-\Delta e/\Delta \log p$ along the loading paths (Fig. 4), which corresponds to the slope at the inflection point on the measured compression curve. It is hard to use a unitary compression index to illustrate the behavior of compression of the compacted tailings, as the *e*-log*p* curve is nonlinear along the loading path. Thus, the compression index is determined in stages and indicated as C_c^* (Deng et al. 2012; Ye et al. 2014).

As shown in Fig. 6, when the pressure is low, the change in the compression index is small, indicating that the soil grain maintains its original state very well. When the pressure exceeds a certain value, the compression index changes significantly, indicating that the structure of the tailings is destroyed (Richard et al. 2010). The trend of compression indexes indicates that the tailings structure was affected by the Cu^{2+}. The compression index of the tailings saturated with the $CuSO_4$ solution was larger than the tailings saturated with distilled water. The influence of the $CuSO_4$ on

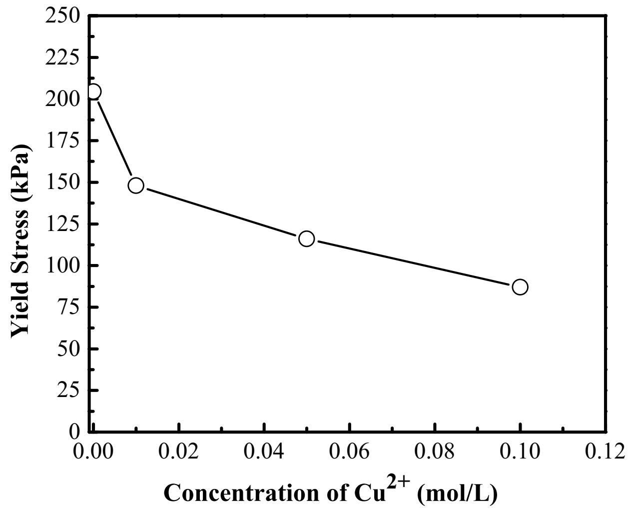

Fig. 5 Yield stress of compacted tailings with various concentrations of $CuSO_4$

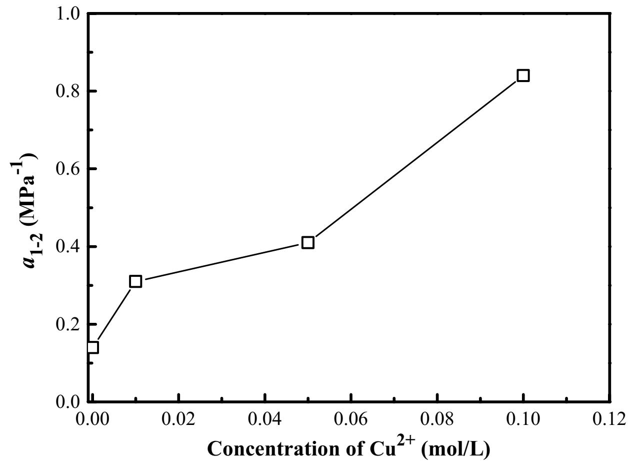

Fig. 7 Effect of concentration of Cu^{2+} on the compression coefficient of the compacted tailings

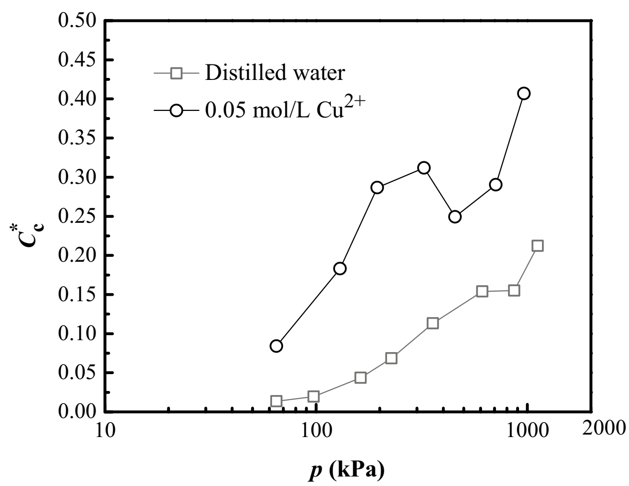

Fig. 6 Compression indexes of compacted tailings with distilled water or $CuSO_4$

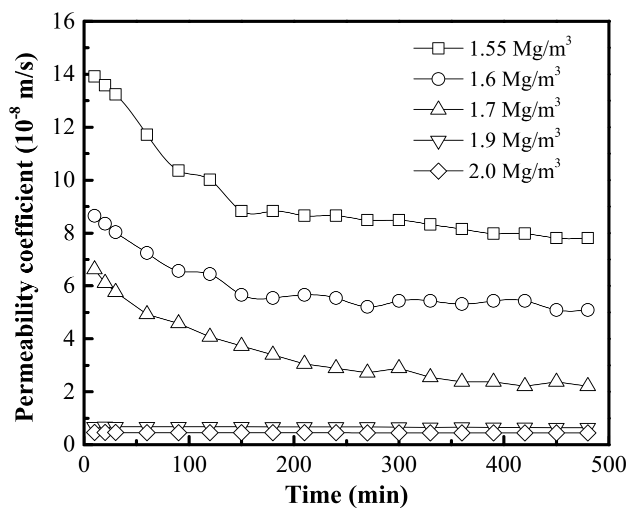

Fig. 8 Permeability coefficient versus time of compacted tailings with various dry densities

the compressibility is indicated by the difference between the two samples.

The effect of Cu^{2+} on the compression coefficient of the tailings is shown in Fig. 7. The compression coefficient of the tailings increased gradually from 0.14 to 0.84 MPa^{-1} as the $CuSO_4$ concentration increased. Furthermore, the character of the tailings changed from a medium- to a high-compressibility soil. The samples had strong compressibility during the initial stage of the consolidation test in distilled water, and then compressibility decreased slowly. Moreover, due to the infiltration of metal ions into the tailings, the tailings disaggregated, enhancing the compression deformation of the tailings and changing the compression coefficient dramatically.

Permeability of the Compacted Tailings

Effect of Dry Density on the Permeability of Tailings

The evolution of the permeability coefficient of tailings with various dry densities is shown in Fig. 8. The curve of the permeability coefficient change with time shows that the permeability coefficient decreased rapidly as the tailings became saturated. Because of the rapid increase in the tailings saturation, the seepage path becomes more complicated, and the permeability coefficient continues to decrease while the permeability coefficient tends to be steady. The value of the permeability coefficient of the tailings decreases gradually with an increase in the dry density. Moreover, the stable permeability coefficient of the tailings decreases gradually. In addition, the fitting line of the permeability coefficient to void ratio is shown; the linear relationship between the permeability coefficient and void ratio can be described by $k = -6.48 + 17.17e$ (Fig. 9). The dry density affected the permeability of the tailings because of the change in porosity. The dry density increased and the void ratio of the tailings decreased as the pores filled with clay, which decreased the porosity and permeability coefficient (Yin et al. 2007).

Effect of Cu^{2+} Concentration on the Permeability Coefficient of Tailings

Figure 10 shows that the permeability coefficient of tailings increased stably with increasing $CuSO_4$ concentrations. Moreover, the permeability of the tailings was increased by the Cu^{2+}; the final stable permeability coefficient was 8.15×10^{-8} m/s when the $CuSO_4$ concentration was 0.05 mol/L and 7.29×10^{-8} m/s when the $CuSO_4$ concentration was 0.01 mol/L. However, the stable permeability coefficient of the tailings in distilled water was only 5.21×10^{-8} m/s, indicating that Cu^{2+} evidently enhanced the permeability of the tailings. The infiltration of the metal contaminant evidently destroyed the connection between grains, changed the pattern of the pore arrangement, and increased the number of pores, thereby lessening the connection between grains. Thus, the permeability coefficient of the tailings increased gradually (Liu et al. 2013).

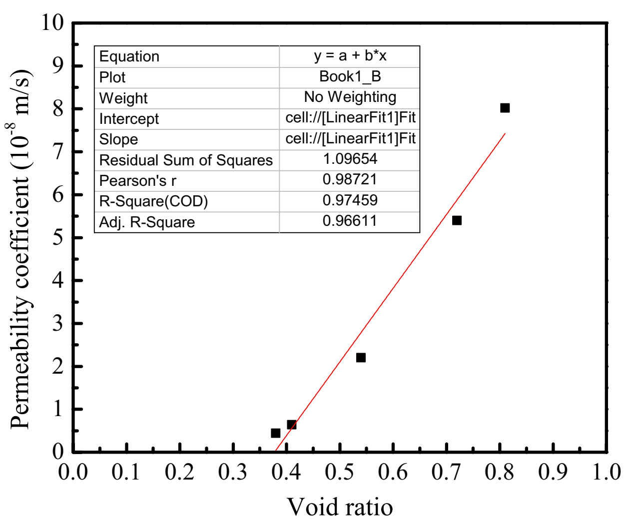

Fig. 9 Fitted results of the relationship between the permeability coefficient and void ratio

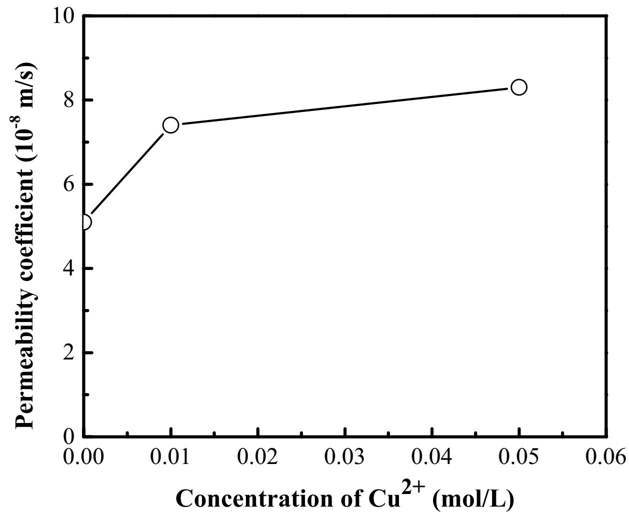

Fig. 10 Variation in the permeability of tailings with respect to the concentration of $CuSO_4$

Effect of Cu^{2+} on the Microstructural Behavior of Tailings

The grain size distributions of the tailings before and after the test are shown in Fig. 11 and Table 3. The content of fine-grained soil in the tailings decreased from 87.38 to 23.96% after the hydromechanical test, whereas the average particle size increased. The mineral composition analysis show that the tailings contain some clay (Supplemental Figure S-3). According to the diffusion double layer theory, the diffusion double layer on the clay surface of tailings was compressed, and the surface potential decreased because of the Cu^{2+} absorbed by the negatively charged tailings. This process decreases the mechanical strength of the tailings

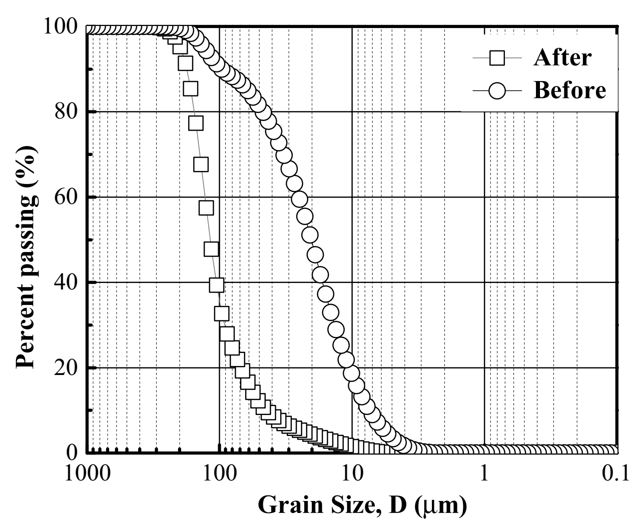

Fig. 11 Grain size distribution of tailings before and after testing

Table 3 Main physical properties of tailings before and after the experiments

Tailings	d_{60}/mm	d_{10}/mm	C_u	C_c	Fine-grained soil/%
Before	24.80	7.56	3.28	0.98	87.38
After	126.72	42.61	2.97	1.19	23.96

(Laine and Karttunen 2010). Moreover, during the migration of Cu^{2+}, a series of redox reactions occurred, which covered the tailings particles or bound adjacent particles together, thereby increasing the tailings' particle size (Fig. 11). Therefore, the Cu^{2+} enhances the compressibility, permeability, and migration of metal contaminants in the tailings (Wu et al. 2017b), which is consistent with the results mentioned above.

Conclusions

We used a custom-designed apparatus to study the influence of Cu^{2+} on the compressibility and permeability of tailings sampled from a copper mine in Jiangxi Province, China. Next, we compared the compressibility and permeability of tailings with different dry densities with various concentrations of $CuSO_4$ solution infiltration. The main observations and conclusions were:

1) The Cu^{2+} affected the compressibility and framework of the tailings and disaggregated the tailings particles, making the structure loose, which led to increased tailings compressibility. At the same dry density, the Cu^{2+} increased the porosity and compressibility of the tailings samples. Moreover, the greater the concentration, the greater the compressibility index.
2) Changes in the dry density of the tailings affected the permeability coefficient of the tailings. As the dry density increased, the permeability coefficient of the tailings gradually decreased. Furthermore, a linear relationship between the permeability coefficient and void ratio was given by $k = -6.48 + 17.17e$.
3) The infiltration of Cu^{2+} compressed the diffusion double layer on the clay surface of the tailings and decreased the surface potential, thus enhancing the compressibility and permeability of the tailings.

Acknowledgements The authors thank the National Natural Science Foundation of China (Project 41807253) and the Natural Science Foundation of Hunan Province, China (Project 2019JJ50763) for their financial support. The authors also thank the Research Fund Program of the Key Laboratory of Geotechnical and Underground Engineering (Tongji University), Ministry of Education (Project KLE-TJGE-B1803).

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