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Geotechnical and Geochemical Assessment of Mine Wastes from Sabah, Malaysia for Biocementation Improvement

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Abstract. Mining wastes are known to be harmful to the environment especially when unregulated, untreated, or abandoned. The first step in their remedial action is the characterization of the geotechnical properties. The aim of this research is to investigate the properties of two Copper mine wastes obtained from Lohan and Bongkud in Sabah, Malaysia, for potential biocementation treatment. The methodology includes soil classification, determination of engineering properties, geochemical properties, and microstructure fabric. Classification results indicated that while both soils can be classified as SM and A-4 according to USCS and AASHTO respectively, Lohan waste has higher coarse-grained particles (69.8%) compared to Bongkud (58.1%). Both soils have a low liquid limit (LL<50), low plasticity (PI <7) and low liquidity (LI<1), and a close range of specific gravity (2.65-2.71). However, Bongkud has higher pH (5.27) and natural moisture content (28.14%) due to its higher organic content (1.27%). In terms of engineering properties, Lohan and Bongkud have MDD at 1640 and 1700 kg/m^3 and OWC at 16 and 15%, respectively, with low cohesiveness and a high angle of friction. With hydraulic conductivity, *k* falls within the range of 10^{-2} cm/s, and these soils are classified as having a medium degree of permeability. Geochemical analysis indicated the presence of nine heavy metal elements with Pb (0.535 mg/L) and Ni (1.092 mg/L) exceeded the safety level in Lohan and Bongkud, respectively. SEM analysis shows both soils have a high degree of disorientation. In conclusion, both soils can be benefitted from biocementation treatment due to the SM classification, medium degree of permeability, and high heavy-metal contaminations.

Keywords: Copper mine waste; Geotechnical properties; Geochemical properties; Microstructure fabric; Mine waste

1. Introduction

Land contamination commonly occurs in agriculture and manufacturing industries However, geotechnical research over the last few decades has focused more on the mining industry, due to the significant increase in global demands for minerals and metals (Hu *et al*., 2017). The geotechnical properties of heavy metal-contaminated soil must be known in order to promote their uses and remediation. Geotechnical evaluation and characterization of soil and landfill wastes were performed prior to the installation of the solar panel (Annavarapu *et al*., 2009) assessment for remediation mitigation by biocementation (Soon *et al*., 2013), analysis of landfill stability (Bahsan *et al*., 2017) and usage as synthetic material (James, Lakshmi, and Pandian, 2017).

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Meanwhile, mineralogical and geochemical evaluation of mine wastes were performed for environmental impact protection assessment (Assawincharoenkij *et al*., 2018; Sracek *et al*., 2010; Kim *et al*., 2007), soil stabilization, (Musta and Kassim, 2000), contamination migration, (Jelenová *et al*., 2018; Mohammad Ali *et al*., 2014) and water quality (Sracek *et al*., 2004).
Previous improvement of contaminated soil, including physical repair, is considered costly for the small-scale project, and chemical passivation via reduction of the migration ability of the heavy metals has been attempted. However, the long-term durability of these methods is still arguable (Mujah, Shahin, and Cheng, 2017). Biological remediation, including phytoremediation and animal repair, was also tried. At the same time, both methods show the low-cost advantage, limited heavy metal enrichment, and slow and long treatment cycles are considered disadvantages (Luo, 2019), thus the emerging popularity of sustainable treatment methods using biocementation.

Prior to biocementation treatment, the soil is characterized to assess its suitability based on selected criteria i.e. medium to a high degree of permeability in the range of 10^{-3}-10^{-4} m/s and containing the amount of heavy metal content exceeding the Safety Standard. In addition, the percentage of gravel, sand, and silt must be feasible to bridge $CaCO_3$ contacts between coarse aggregate particles (Mahawish, Bouazza, and Gates, 2017). As observed in a study of waste soils, lower dry density can generally be equated to a lower strength, higher permeability, and worse volume stability (Dhir, Ghataora, and Lynn, 2016). Additionally, contamination reduces the shear strength of soil, as reported by Ratnaweera, and Meegoda, (2006). Hence pre-treated values and initial microstructure fabric are to be determined to confirm the literature and to be used as comparison upon treatment.

Therefore, the objective of the research is to characterize the geotechnical properties of two Copper mine wastes, including their soil classification, degree of permeability, shear strength parameters, and compaction. Additionally, the study investigates the geochemical properties, such as heavy metal contaminants and microstructure fabric.

2. Methods

Soil sampling of mining wastes was conducted in two Lohan and Bongkud, near Mamut, District of Ranau, Sabah, by the trial pit. Mamut Copper Mine, which was in operation from 1975 to 1999, generated about 350 Mt of waste (van Der Ent and Edraki, 2016). The wastes from this mine were then dumped at Lohan and Bongkud. The samples were collected at a depth of 0.5 m to 3.0 m of the soil profile according to BS10175:2001, which regulates subsurface investigation not exceeding 4.5m. The sampling location for Lohan and Bongkud was an approximate GPS of 6° 0' 45.936'' N, 116° 44' 20.004'' E and 6° 1' 20.6" N, 116° 44' 28.1" E, respectively. The Lohan point was selected within the perimeter of abandoned dam facilities, while the Bongkud was selected at a slope of 2.5% towards the north of Lohan, the area where seepage and discharge from the Lohan dam could flow into the nearby village land. A total of 3 sample batches were collected from each sampling point. Collected samples were prepared according to BS 5930-2015. They were carefully removed from the sampling spot, with no extraneous water shall come into contact. In addition, the sample was protected from sunshine and winds and covered. No prior treatments were made to the soil before testing.

2.1. Soil Classification and Characterization

Soil classification and characterization were performed according to BS 1377 procedures. The specific gravity was determined by ASTM D854 using a pycnometer bottle, while organic content was determined using ASTM D2974. A microprocessor-based benchtop pH (Model Therom Scientific Stara 1117) that can measure pH while compensating for the effects of temperature was used for the acidity and alkalinity study.

2.2. Geotechnical Engineering Properties

Standard Proctor compaction tests were conducted to determine the maximum dry density achievable for a specific soil type by applying controlled compactive force at an optimum water content. Additionally, direct shear tests were performed to evaluate the shear strength parameters of the mining waste soils. Meanwhile, the hydraulic conductivity, (k) of a mine waste soil was determined by the constant head test method. All the procedures complied with BS1377.

2.3. Geochemical Properties

Inductively Coupled Plasma-Optical Emission Spectrometer (ICP-OES) using the Rigoku SmartLab® model was used to detect the heavy metal elements level because it is highly sensitive and can detect beyond mg/L and quicker analysis time for about 10 minutes per sample per element.

2.4. Microstructure Fabric

Scanning Electron Microscopy, or SEM analysis using the Hitachi SU8020 model, was used to provide high-resolution images of soil samples for evaluation of their surface flaws, fractures, and corrosion.

3. Results and Discussion

3.1. Soil Index Properties

Laboratory soil samples from both locations are presented in Table 1. Both Lohan and Bongkud are predominantly coarse-grained soils with 69.8 % and 58.1 % and predominantly sand (55.1 %, 44.8 %), respectively. Significant amounts of fine-grained soil, particularly of silt (19.2 %, 36.4 %), are also observed in both soils. The coefficient of uniformity, Cu, of both of these soils is 60 and 77.8, respectively, while the coefficient of curvature, Cc, of both of these soils is 0.42 and 0.46, respectively, indicating that both the soil is uniform, sometimes referred to as poorly graded. Nevertheless, since the percentage of soil finer than No 200 (0.075mm) is 19.2 % and 36.6% (larger than 5%), these two parameters have little or no meaning (Liu and Evett, 2014).

Table 1 Classification of Copper Mine Wastes from Sabah, Malaysia

Properties	Lohan Dam	Bongkud
Coefficient of uniformity	60	77.78
Coefficient of curvature	0.42	0.06
Particle size distribution (%)		
Coarse	69.8	58.1
Gravel	14.7	9.7
Sand	55.1	48.4
Fine	30.2	41.9
Silt	19.2	36.4
Clay	11.0	5.5
Classification		
USCS	SM	SM
AASHTO	A-4	A-4

Therefore, it can be concluded that based on classification by Unified Soil Classification System (USCS), they are classified as SM and A-4, a fair to poor rate as road subgrade according to the American Association of State Highway and Transport Officials (AASHTO).

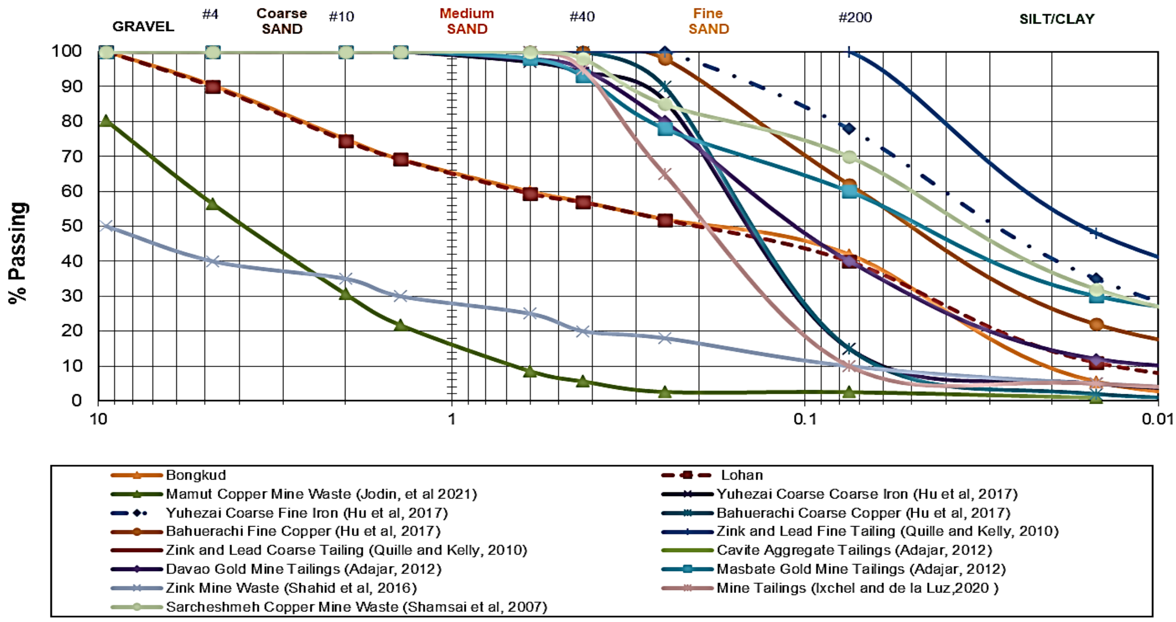

Figure 1 Particle Size Distribution of Lohan and Bongkud Compared to Other Mine Wastes

Comparison between their particle size distribution to copper and iron mine wastes (Hu *et al*., 2017), zinc and lead tailings (Quille and Kelly, 2010), aggregate and gold mine tailings (Adajar, 2012), zinc mine waste (Shahid *et al*., 2016), mine tailings (Ramos-Hernández and Pérez-Rea, 2020) and copper mine waste (Shamsai *et al*., 2007) are shown in Figure 1. On average, copper tailings from Lohan and Bongkud are relatively coarser. However, these properties could depend on the milling size, feeding materials, hydro cyclone pressure, slurry density, and presence of water during the extraction process (Shamsai *et al*., 2007). However, compared to the previous study of Mamut mine wastes (Makinda *et al*., 2021) and zinc mine waste (Shahid *et al*., 2016), the percentage of coarse-grained particles is generally lower in Lohan and Bongkud because the mining process in Mamut include drilling, blasting, loading, and hauling, while the ore extraction process includes crushing, grounding and floating (Azizli *et al*., 1995).

As shown in Table 2, both Lohan and Bongkud can be classified as having low liquid limit (<50%) observed by the similar value of LL at 27 %. Lohan recorded PL at 23.5% and Bongkud at 22.6%, indicating almost similar amount of water content needed to change their phase from plastic to semi-solid state. Both Lohan (PI = 3.5) and Bongkud (PI = 4.4) can be considered as having low plasticity (PI < 7) due to the presence of silty content in the soil. The low values of the liquidity index (LI) for both soils indicate a dense relative density at their natural water content. According to the soil activity classification, the Lohan soil (A = 0.32) is considered inactive, while the Bongkud soil (A = 0.80) is classified as normal. Range of specific gravity for Lohan is 2.66 - 2.70, almost similar to Bongkud at 2.65-2.71. As the specific gravity depends upon the materials that made the weight of soil, it can be concluded that Lohan and Bongkud contained similar material and heavy metal elements. (Islam, 2021). The specific gravity could also be affected by the organic matter and particles in the soil volume. In subsequent tests, it can be observed that the organic content of Lohan is 0.77% and Bongkud is 1.27%.

Table 2 Characterization of Copper Mine Wastes from Sabah, Malaysia

Properties	Lohan Dam	Bongkud
Consistency		
Liquid Limit, LL (%)	27.0	27.0
Plastic Limit, PL (%)	23.5	22.6
Plasticity Index, PI (%)	3.5	4.4
Liquidity Index, LI (%)	0.91	-0.79
Activity	0.32	0.80
Average Specific Gravity, Gs	2.66−2.70	2.65−2.71
Average Organic content, (%)	0.77	1.27
Average Natural Moisture, (%)	26.68	28.14
Average pH	6.18	5.27

The organic content is below 30% for both sites, the minimum amount suggested by United Nations Food and Agriculture Organization for cropping soils. The organic content was observed to affect the characteristics of copper mine wastes from Zambia (Chileshe *et al*., 2020). Lohan has higher natural moisture (26.68%) than Bongkud (19.14%), while Bongkud has a higher amount of fine-grained (41.9%) than Lohan (30.2%). The nature the mine is evident in the pH value recorded at 5.27 for Bongkud and Lohan (6.18). Previous research in 2008 in the Mamut area focusing on water quality discovered a pH range of 2.90 – 3.75. The acidic nature was attributed to the high and variable total acidity (176 – 1697 mg $CaCO_3$ /L), high TDS (302 – 2673 mg/L), high sulfate (292 – 2808 mg/L), and elevated concentrations of dissolved metals; Al, Mn, Fe, Cu, and Zn (Jopony and Tongkul, 2009).

3.2. Geomechanical Properties

3.2.1 Compaction Properties

Figure 2 shows the selected compaction tests to represent the highest, medium, and lowest curves of each soil.

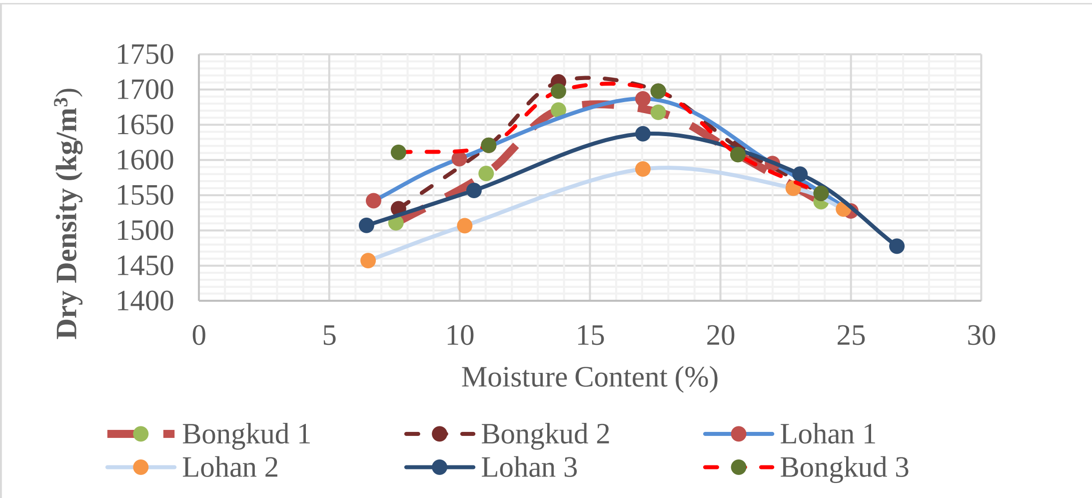

Figure 2 Compaction Properties of Lohan and Bongkud

The tests yielded a range of MDD between 1587– 1685 and 1670 – 1710 kg/m^{3,} with an average of 1640 and 1700 kg/m^3 for Lohan and Bongkud, respectively. By comparison, a previous study on an Iran copper mine achieved MDD of 1780 kg/m^3 (Shamsai *et al*.,2017), while copper mines in in several regions of Bulgaria were found to have within the range of 1317 –1775 kg/m^3 (Germanov, 2003). Meanwhile, the ranges of OWC achieved were between 15–17 and 14.5–15.5 %, with an average of 16 and 15 % for Lohan and Bongkud, respectively. The Optimum Water Content (OWC) represents the moisture content at which a specific compactive energy produces the highest dry density, thereby improving the soil properties. Soils with higher OWC values, like Lohan and Bongkud, require a greater amount of water to achieve this optimum level.

3.2.2 Shear Strength Parameters

The shear strength parameters shown in Table 3 show that mine waste soils exhibit low cohesion values, ranging from 26 – 34 kPa for Lohan and Bongkud (25 – 35 kPa). By comparison, the cohesion of dry copper mine soil varies from 0.07 kg/m^2 to 0.15 kg/m^2 (Shamsai *et al.*, 2007). Similar findings were observed in coarse-grained Yuhezhai iron tailings and Bahuerachi copper tailings (Hu *et al.*, 2017).

Table 3 Shear Strength Parameters of Copper Mine Wastes from Sabah, Borneo

Shear Strength Parameters	Lohan	Bongkud
Range of Angle of Friction (°),	13.6 – 18.0	12.5–17.5
Range of Cohesion (kPa)	26 – 34	25 – 35

On the contrary, the range angle of friction of Lohan is 13.6 – 18.0° and 12.5 –17.5° for Bongkud. By comparison, the friction angle in drained tests was found in the range of 12.5–18.3°, and for undrained tests was found at 16.0–20.4° for sulphide-rich tailings (Bhanbhro, 2014). The higher angle of friction of unprocessed tailings is because of their high angularity compared to natural granular materials.

3.2.3. Permeability

The average permeability of the three copper mines from Sabah is shown in Table 4, with Lohan's recorded value of 3.77 x 10^{-2} cm/s and Bongkud at 3.51 x 10^{-2} cm/s. Both soils fall within a 'medium' degree of permeability, with *k* ranging from 10^{-3} to 10^{-1} cm/s corresponding to the 'clean sand and gravel mixture' soil type. The higher degree of permeability in all soils can be attributed to the higher void ratio in mine waste soil (Hu *et al.*, 2017).

Table 4 Permeability of Copper Mine Wastes from Sabah, Borneo

Properties	Lohan	Bongkud
Average Permeability (cm/s)	3.77 x 10^{-2}	3.51x 10^{-2}

Biocementation treatment by EICP is proven to be effective in reducing hydraulic conductivity. The permeability decreases by 92% on the first injection and a further 73% by the second injection for a total reduction of 98% (Nemati and Voordouw, 2003). Treatment was performed with 12 g/L urea and 30 g/L of $CaCl_2$ with Jack bean urease-based enzyme and addictive. Another research using 36 g/L of urea and 90 g/L of $CaCl_2$, and 0.3 g/L Jack bean urease based without addiction found 62% in hydraulic conductivity reduction (Nemati, Greene, and Voordouw, 2005).

The effectiveness of the treatment is a result of $CaCO_3$ crystals that fill the space between soil particles change the soil pore volume, and hence reduce soil hydraulic conductivity (Almajed *et al.*, 2021).

3.3. Geochemical Properties

Table 5 shows that level of the heavy metal element Bongkud is lower than Lohan except for Cr, Mn, and Ni. The high level of iron (Fe) abundantly in Lohan soil is derived from a wide variety of rock weathering under extreme leaching and humid oxidizing conditions.

Table 5 Heavy Metals Element in Lohan and Bongkud

Heavy Metal Elements	Wavelength	Average Content(mg/L)	
		Lohan	Bongkud
Arsenic, As	193.696	0.319	0.169
Cadmium, Cd	226.502	-0.051	-0.058
Cobalt, Co	230.786	-0.025	0.008
Chromium, Cr	267.716	0.586	2.787
Copper, Cu	324.754	4.337	1.846
Iron, Fe	238.204	2931.4	2133.80
Manganese, Mn	257.610	3.642	5.303
Nickel, Ni	230.299	0.376	1.092
Lead, Pb	405.781	0.535	0.274
Zinc, Zn	213.857	0.555	0.140

Comparing the level of heavy metal elements to the standard of the Department of Environmental Malaysia (DOE, 2014) and the Food and Agriculture Organization of the United Nations (FOA) for residential soil, the concentration of Nickel, Ni in Bongkud was found to be beyond the safe level of 0. 41 mg/L, while the concentration of Lead, Pb in Lohan was beyond the safety level of 0.3 mg/L. Subsequent comparisons made to the safety level for groundwater indicate that the As, Cr, Cu, Ni, Pb, and Zn levels in both soils are above the safety limit, while Co exceeded the safety limit in Bongkud only.

3.4. Microstructure Fabric

Both Lohan and Bongkud soil shows a higher degree of disorientation, as shown in Figure 3. Comparison of before and after in SEM images can also be used to indicate the effect of treatment or stabilization as newly formed bonding or materials may cover the surfaces of the sample (Ural, 2021) An example can be observed in the biocementation of treated residual tropical soil (Soon *et al.*, 2013). Bladelike form was observed and later identified as calcite.

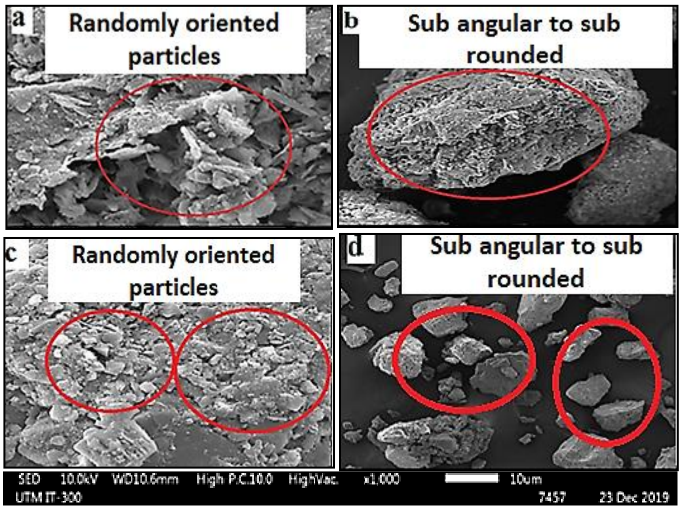

Figure 3 (a-b) Lohan and (c-d) Bongkud Microstructure Fabric

The SEM analysis appears to be composed of powdered and hardened particles, and the soil particle arrangement can be classified as 'randomly oriented particles between two domains' as parallel domains are randomly interrupted by smaller soil particles. A closer inspection of the soil surface indicates a high degree of angularity. By size comparison, Lohan soil shows a higher degree of disorientation compared to Bongkud. Their surfaces appear to be very smooth, with no agglomeration between particles. This may indicate that, without treatment, such as biocementation, the strength value of mine waste is low due to the low bonding between the loose grain structure.

4. Conclusions

In summary, the study describes the characteristics of Lohan and Bongkud copper mine wastes, such as their soil classification, degree of permeability, shear strength parameters, compaction, heavy metal contamination, and microstructure fabric. Both mine wastes were discovered to have a high potential for environmental contamination due to their medium degree of permeability, low cohesiveness, high angle of friction, and high levels of heavy metals. The severe soil pH situation is expected to make any attempt at phytoremediation expensive. Meanwhile, both soils are suitable and will benefit from bioremediation treatment based on the previously listed criteria. Due to the soil heterogeneity, future research could benefit from a larger sample size to ensure accurate representative of of the mine waste soils.

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