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AN INVESTIGATION ON THE USE OF T'BOLI GOLD TAILINGS FROM MINDANAO STATE UNIVERSITY – ILIGAN INSTITUTE OF TECHNOLOGY (MSU-IIT) GOLD PILOT PLANT AS TOTAL REPLACEMENT OF FINE AGGREGATES IN CEMENT MORTAR

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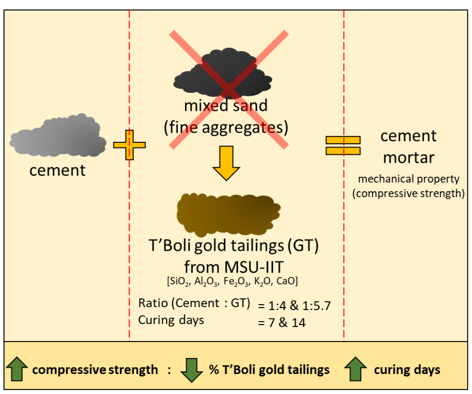
Graphical abstract



Abstract

Gold tailings are waste products produced from the extraction and recovery of gold from its ore. At present, these tailings are in large amounts and are stored in tailings storage facilities, mostly in the form of dams. These gold tailings dams could have serious consequences for the environment and humans if unfortunate events happen. Thus, it is important to utilize these wastes not only to give them value but also to lessen the environmental and human risks they bear. This study investigates the use of gold tailings produced by the Mindanao State University – Iligan Institute of Technology (MSU-IIT) gold pilot plant from the gold ore of T'Boli, South Cotabato, Philippines, as a total replacement of the sand (fine aggregates) component in cement mortar. The chemical compositions of the T'Boli gold tailings were identified using XRF and SEM-EDX analyses. It was found that SiO_2, Al_2O_3, Fe_2O_3, K_2O, and CaO were the major oxide minerals found in the tailings sample. To investigate the effect of the gold tailings on the mechanical property (compressive strength) of the cement mortar, two different formulations of cement and gold tailings (20:80 and 15:85), a controlled formulation with no gold tailings, and two different curing periods (7 and 14 days) were used. The result shows that the formulation of the control sample produces the highest compressive strength of 20.95 MPa at a 14-day curing period. This is followed by the cement mortar in ratios 1:4 and 1:5.7 at a 14-day curing period with a compressive strength of 9.11 MPa and 7.14 MPa, respectively. With these findings, it shows the suitability of T'Boli gold tailings (mining waste) as an alternative to fine aggregates in cement mortar.

Keywords: gold tailings, cement mortar, compressive strength

1.0 INTRODUCTION

Different minerals and metals that are essential to the growth of society are extracted from their primary sources, which are their ores. As the demand for these minerals and metals increases, there is also a need for increased mining of their ores. The fact that ore grades are now lower, the scale of ore processing is expanding. As a result, tailings or mining waste will be produced in greater quantities in the near future. In fact, in 2010, approximately 14 billion metric tons of tailings were produced by the mining industry globally [1].

Gold tailings (GT) are wastes that are generated from the extraction of gold from its ores and are one of the most commonly produced tailings worldwide. Currently, most of the gold processing plants are storing these wastes in tailings storage facilities, or dams, and were regarded as having no economic value. Gold mine tailings facilities or dams are a high-risk mining practice due to the presence of heavy metals (HM) that could enter natural springs and lead to serious human health issues when consumed [2]. Also, unrehabilitated gold tailings take up a lot of space, and can affect air quality due to the formation of dust that may cause serious human respiratory health problems [3]. Thus, improved gold tailings management, like recycling, should be practiced to lessen the negative effects of these tailings.

The Mindanao State University - Iligan Institute of Technology (MSU-IIT) gold pilot plant has been producing gold tailings as a by-product of the gold recovery operations. The aim of the pilot plant is to physically separate the gold from its ore with the use of a comminution process and a gravity separator. Unlike any other gold tailings produced by the gold industry in the Philippines, the gold tailings in the MSU-IIT gold pilot plant were not produced with the aid of different toxic chemicals like cyanide and mercury. Thus, it is expected that there will be no cyanide or mercury in its gold tailings, and this makes the gold pilot plant tailings unique compared to any gold tailings from the different gold-producing plants in the Philippines.

Even though there is an absence of cyanide and mercury in the MSU-IIT gold tailings, the oxidation of pyrite (FeS_2) in the tailings can trigger acid mine drainage (AMD) that could pollute the water [4]. Also, when these tailings are in large volumes, they may take up a lot of space in the facility. Therefore, there is still a need to utilize this waste, like using it as a component in developing different construction materials.

Construction materials like concrete blocks and cement mortars are extensively used now [5]. It is used in the construction of commercial and residential buildings, internal partition walls and boundary walls, architectural decoration, and the finishing of concrete walls. And with the increasing population worldwide, there will also be an increasing demand for shelter. As a result, there will be an increase in demand for construction materials like cement mortar.

Research is now into the production of sustainable construction materials. These are mostly produced by incorporating different wastes as a major component of these construction materials. Wastes like recycled glass, fly ash, foundry sand, copper slag, and tailings are some of the wastes incorporated into the development of concrete blocks and cement mortars [6, 7, 8, 9, 10, 11, 12, 13, 14]. However, with the adequate silica content of gold tailings, it has the potencial to be a replacement for mixed sand (fine aggregates) used in common concrete blocks and cement mortars.

The development of cement mortars incorporating gold tailings has already been investigated by researchers. But there have been no investigations into the use of gold tailings produced from a chemical-free processing line in the development of cement mortar. Here, the effect of MSU-IIT gold pilot plant tailings on the mechanical property (compressive strength) of the cement mortar will be investigated.

2.0 METHODOLOGY

2.1 Materials

The study is using ordinary Portland cement purchased locally and gold tailings from the MSU-IIT gold pilot plant in Brgy. Hinaplanon, Iligan City, Lanao del Norte, Philippines. Figure 1 shows the particle size distribution of the T'Boli gold tailings, which shows 58.20% passing an 80-mesh sieve (180 µm). Ordinary mixed sand obtained locally in Iligan City was used as a control in the experiment. Normal tap water was used for the cement mortar component and curing. Lastly, a customized metallic molder was used to mold the cement mortar samples.

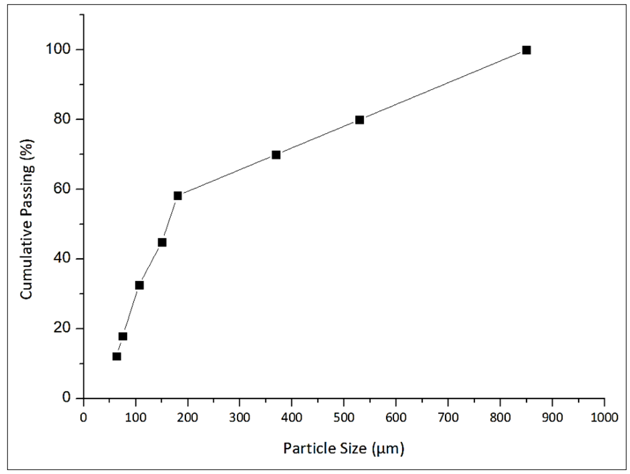


Figure 1. Particle size analysis (PSA) of T'Boli Gold Tailings

Portland cement has a specific gravity that ranges from 3.15 to 3.16 [8, 10] while ordinary sand (fine aggregates) has a specific gravity that ranges from 2.54 [8] to 2.65 [15]. Which shows that ordinary Portland cement is relatively denser than ordinary sand. Sand used for cement mortar preferably has a particle size distribution of 10% to 30% passing a 50-mesh sieve and 2% to 10% passing a 100-mesh sieve [15]. While an ordinary Portland cement particle's size typically ranges from 1–50 µm [16].

2.2 Chemical Characterization of the Gold Tailings

X-ray fluorescence (XRF) analysis and scanning electron microscopy – energy dispersive X-ray (SEM-EDX) spectroscopy were used to determine the chemical composition of T'Boli gold tailings samples. Representatives of 50 grams were obtained using the standard sampling methods. Then, these samples were kept in sealed bags and sent to Hokkaido University, Japan.

2.3 Cement Mortar Mix Design and Preparation

The formulations for the experiment were based on the standard formulation for making cement mortars. The standard formulation is 20% fine aggregates (mixed sand) and 80% cement, or a 1:4 ratio [17]. Other set-ups were operated where mixed sand was totally replaced by gold tailings: 20% and 80% (1:4 ratio) and 15% and 85% (1:5.7 ratio), respectively.

It was found that the 1:3 ratio is the optimum ratio for cement and sand in cement mortar production [18], which is predictable since the higher the binder proportion, the higher the compressive strength of the cement mortar. However, the 1:4 ratio has been used as a standard for the production of cement mortar, which is equal to 20% cement and 80% sand.

This study would like to reduce the use of cement (the binder) by 5% to observe its effect on the compressive strength of the cement-based mortar with T'Boli gold tailings. Thus, making the decision to use the formulation of 15% cement and 85% sand, which is equal to a 1:5.7 ratio, Table 1 shows the different setups used in the study. Moreover, three replicates were performed in each setup.

Table 1. Mix proportions used in the experiment.

Mix ID	Ratio		
	Cement	**Mixed Sand**	**Gold Tailings**
CM01	1	4	0
CMO2	1	0	4
CM03	1	0	5.7

The raw materials for mixing and molding were mixed sand, T'Boli gold tailings, Portland cement, and water. These materials, in predetermined amounts, were thoroughly mixed and dumped into the customized metallic molder of 2x2x4 inches to produce cuboid specimens, which is the standard size required for the mechanical testing in the testing laboratory. Pressure was applied to compact the mixture to avoid the formation of air pockets. After that, the molder was removed, and the molded samples were allowed to dry. The samples were air-dried for 24 hours before the start of curing. In the curing, two curing periods of 7 and 14 days were used in the experiment, with three replicates in each set-up. Portland cement develops in 14 days [15] and this study would like to see and investigate the mechanical strength of cement-based mortar with T'Boli gold tailings within 14 days of curing only. The samples were cured in water tank at room temperature in a secured location and were tested after 7 and 14 days. As shown in Table 2, different cement mortar samples produced in different formulations were cured at different curing periods.

Table 2. Curing experimental set-up used in the experiment

Curing Days	Mix ID		
	CMO1	**CM02**	**CM03**
7	CMO1-7	CMO2-7	CMO3-7
14	CM01-14	CM02-14	CM03-14

2.4 Mechanical Property Determination

Compressive strength is a measure of the overall strength of a material under compression. To determine the cement mortar's compressive strength (MPa), the samples were exposed to a controlled compressive force in a universal testing machine. This was done using the standard operating procedure of the City Engineers Office of Iligan City, which is to use cuboid cement mortar specimens. And the compressive strength was determined just after the failure of the sample.

3.0 RESULTS AND DISCUSSION

3.1 Chemical Composition of the Gold Tailings

3.1.1 X-ray Fluorescence (XRF) Analysis

The mineral composition results of the gold tailings by X-ray fluorescence (XRF) are presented in Table 3. It is evident that the T'Boli gold tailings is composed primarily of these oxide minerals, namely: SiO_2, Al_2O_3, Fe_2O_3, K_2O, MgO, Na_2O, CaO, and trace amounts of SO_3, TiO_2, P_2O_5, and MnO. Almost the same chemical composition of gold tailings was observed from the previous studies, with silica, alumina, and iron oxide as the most abundant minerals in the tailings sample, which then suggests that the T'Boli gold tailings have a pozzolanic nature [12, 13, 19, 20, 21, 22, 23, 24]. These T'Boli gold tailings found in storage facilities of the MSU-IIT gold pilot plant tend to clump and harden when exposed to repeated hydration and drying, which could be an indication that its oxides, especially its aluminosilicate materials, could be reactive to the presence of CaO or $Ca(OH)_2$.

The determination of the chemical composition of the gold tailings sample is important because it will determine how qualified the sample is to become an alternative to conventional fine aggregates in the production of cement mortar. Fine aggregates used in the production of cement mortar and cement concrete are usually high in SiO_2, Al_2O_3, and CaO. Commonly, fine aggregates used in concrete and mortar production are composed of SiO_2, Al_2O_3, Fe_2O_3, and CaO [24]. These constituents are essential in the setting and hardening of tricalcium silicate ($3CaO\text{-}SiO_2$) and dicalcium silicate ($2CaO\text{-}SiO_2$), which are also the major constituents of common Portland cement [25]. Aside from SiO_2 and CaO, Portland cement is also composed of Al_2O_3, Fe_2O_3, SO_3, MgO, TiO_2, and K_2O [24]. With all of these, a relatively complicated hydration reaction occurs between the different constituents of the Portland cement, fine aggregates, and water that will harden concretes and mortars. Equation 1 shows one of the most important hydration reactions involving dicalcium silicate [25].

$$2CaO\text{-}SiO_2 + xH_2O = 2CaO\text{-}SiO_2\text{-}xH_2O \qquad \text{(Eq. 1)}$$

Table 3. Chemical composition of the T'boli gold tailings.

Component	Content (% mass)
SiO_2	62.10
Al_2O_3	18.93
Fe_2O_3	7.00
K_2O	5.64
MgO	2.14
Na_2O	1.51
CaO	1.13
SO_3	0.79
TiO_2	0.40
P_2O_5	0.20
MnO	0.14

3.1.2 Scanning Electron Microscopy – Energy Dispersive X-ray (SEM-EDX) Spectroscopy

In the production of Portland cement, materials in some form of calcium carbonate ($CaCO_3$) are essential; thus, it is made from an artificial mixture of lime-bearing materials [15]. Ordinary Portland cement will usually show CaO as the highest overall percentage in its overall chemical composition, followed by SiO_2, Al_2O_3, and Fe_2O_3 [8, 10, and 22]. These oxides can also be found in ordinary sand, but in different proportions, where SiO_2 is the most abundant, followed by Al_2O_3 and Fe_3O_4 [10, 22].

In concrete, the formation of calcium silica hydrates, dicalcium silicate, and tricalcium silicate from the hydration of SiO_2 and CaO is considered to be the main strength contributor of concrete and cement mortar [26]. The higher the SiO_2 and CaO content in a concrete or mortar mixture, the harder the material will be and thus have a relatively high compressive strength.

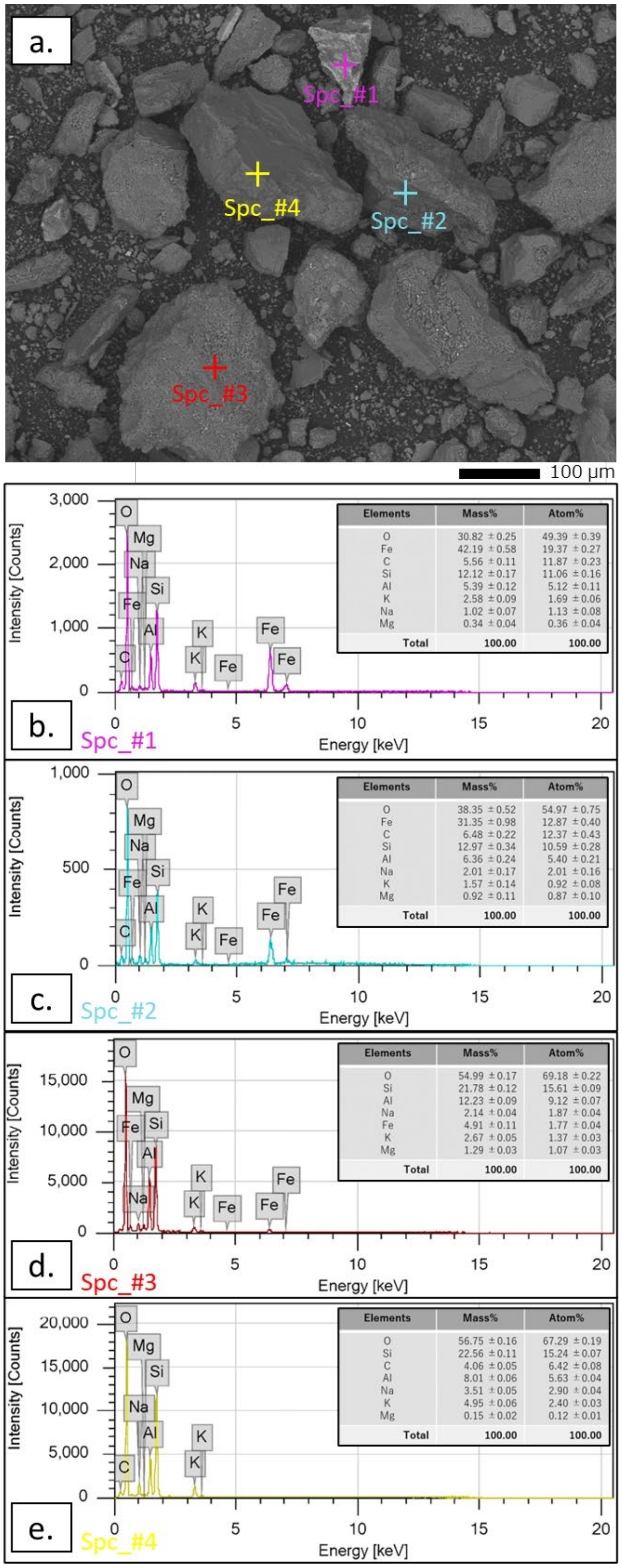


Figure 2. SEM-EDX spectra at different points on the surface of the T'Boli gold tailings: (a) SEM imaging of the T'Boli gold tailings, (b) spectra at point 1, (c) spectra at point 2, (d) spectra at point 3, and (e) spectra at point 4.

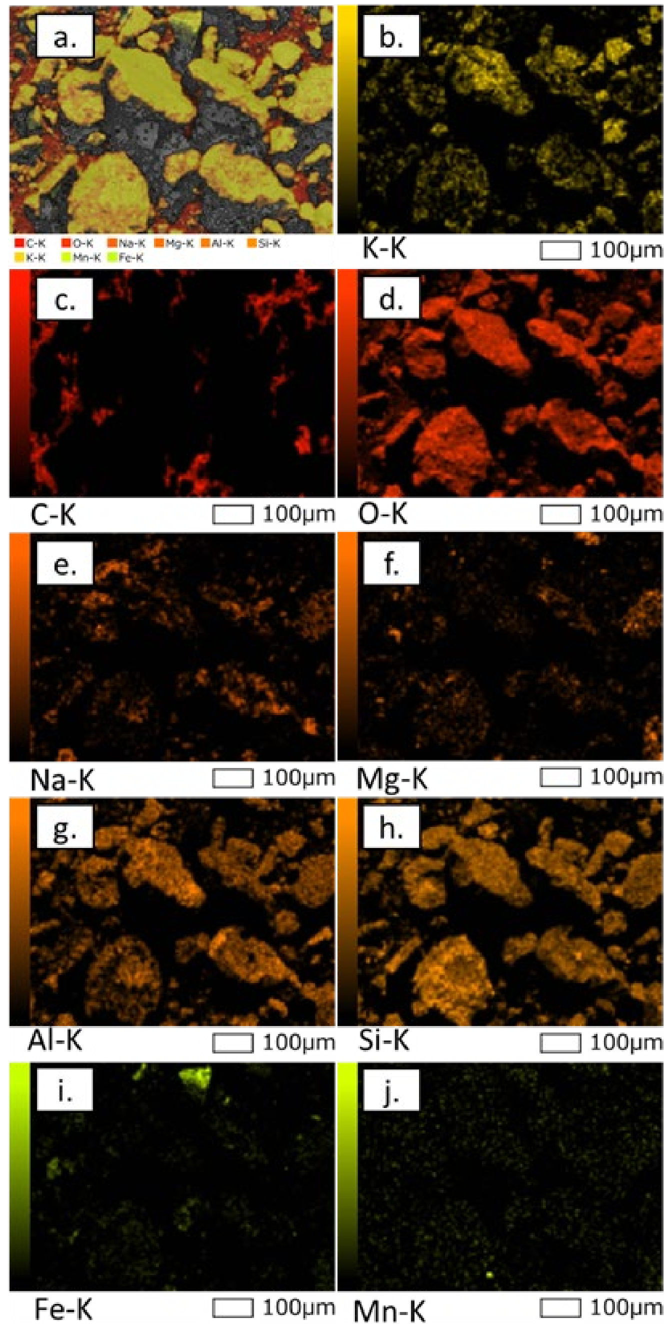


Figure 3. T'Boli gold tailings overall elemental mapping (a), and the corresponding specific elemental mapping of (b) K, (c) C, (d) O, (e) NA, (f) Mg, (g) Al, (h) Si, (i) Fe, and (j) Mn.

While Al_2O_3 could contribute to the compressive strength of the cement mortar, it was found that increasing Al_2O_3 does not give better results in terms of compressive strength. Hence, higher SiO_2 content will contribute more to the improvement of the compressive strength of cement mortar with an adequate amount of CaO than higher Al_2O_3 content in fine aggregates. This is because SiO_2 is one of the main components that could harden concrete and cement mortar through the formation of calcium silica hydrates [27].

The surface structure of T'Boli gold tailings plays an important role in understanding the surface characterization and elemental composition of the samples. The SEM micrograph indicates the broad distribution of finer particles, shows their shapes, and may indicate the porosity of the material. The T'Boli gold tailings consist of irregular-shaped flakes and granular particles. As shown in Figure 2a, SEM confirms that particles of the T'Boli gold tailings have a large crystal morphology and a cracking structure associated with numerous known gold mine tailings properties [22].

Elemental mapping on the surface of the gold tailings was identified using EDX. Figure 2 shows the different elemental abundances in the different areas on the surface of the gold tailings. Each spectrum represents the different points on the surface of the gold tailings where oxygen is shown to be in relatively large abundance. This suggests that the T'Boli gold tailings are mainly composed of oxide minerals. Based on the spectra, these oxide minerals are mainly composed of silicon, iron, aluminum, sodium, potassium, and magnesium.

Figures 2b–2e also show that there are no environmentally regulated toxic and hazardous elements found in the T'Boli gold tailings, like As and Cd, or heavy metals like Cu and Zn. Thus, elemental analysis of gold tailings using SEM-EDX is strongly recommended before these materials are utilized in concrete or cement mortar production [12].

Overall elemental mapping of the T'Boli gold sample is shown in Figure 3a, which shows the relative abundance comparison of the different elements in the T'Boli gold tailings. Each color signal represents how relatively abundant a particular element is in the sample. First, strong signals of O, Al, and Si dominated the mapped region of the sample, as seen in Figures 3d, 3g, and 3h, respectively. This strongly indicates that the major compounds that are found in the gold tailings sample are rich in SiO_2 and Al_2O_3, which are commonly the most abundant compounds in siliceous gold ores [22, 23, and 24]. Additionally, these compounds are also found to be abundant in ordinary sand [10, 21]. Based on chemical composition, T'Boli gold tailings is a suitable substitute for ordinary sand in cement mortar. SiO_2, which is found in abundance in T'Boli gold tailings, will react with CaO from cement during the hydration process to effectively harden the cement mortar.

Meanwhile, other elements like Fe, Mg, Na, Mn, and K were detected, which could be found in Figures 3i, 3f, 3e, 3j, and 3b, respectively. These elements are also commonly found in ordinary sand for cement mortar production. The detection of carbon in figure 3c refers to the carbon tape used during the scanning electron microscopy. These results indicate conformity to the results of the XRF in Table 3. SEM-EDX results imply that the T'Boli gold tailings possess a strong pozzolanic nature and are very compatible as a total replacement of mixed sand (fine aggregates) in developing cement mortar.

3.2 Compressive Strength of the Cement Mortar

The compressive strength of the cement mortars incorporated with different amounts of T'Boli gold tailings was determined using a universal testing machine. As shown in Figure 4, the control sample with no incorporated gold tailings shows the highest compressive strengths of 17.79 MPa and 20.95 MPa from 7 and 14 curing days, respectively. These compressive strengths of a cement mortar are actually much lower compared to the compressive strength predicted from other research to be between 60 and 56 MPa [28]. Sample CM02 (ratio 1:4) shows a compressive strength of 7.46 MPa at the end of 7 days of curing and 9.11 MPa at the end of 14 days of curing. The CM03 sample, with the highest proportion of gold tailings, shows the lowest compressive strengths of 6.31 MPa and 7.14 MPa at the end of the 7- and 14-day curing periods, respectively. However, the current compressive strength of the cement-based mortar with T'Boli gold tailings is possibly much higher if it is investigated after a 28-day curing period.

A simple trend in the changes in the compressive strength of the cement mortar could also be observed in Figure 4. As the proportion of gold tailings increases, there is a decrease in the compressive strength of the cement mortar. This trend is somehow similar to the findings of the study by M. Yildirim Ozen et al. et al. (2020), where their study investigated the suitability of gold tailings as a cement substitute. Moreover, these results are almost identical to the compressive strength of the masonry mortar (after a 7-day curing period) produced by a partial replacement of sand with gold tailings done by S. Vignesh and his colleagues in 2015, where an increasing proportion of gold tailings resulted in decreasing compressive strength.

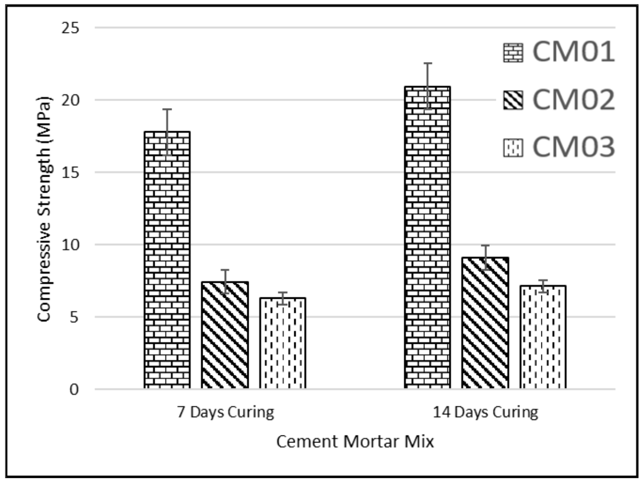


Figure 4. The compressive strength (MPa) of the cement mortar with different proportions of gold tailings at different curing period.

The study of Ince, C. (2019) observed that there is an increase in compressive strength with increasing gold tailings proportions of 10%, 20%, and 30%, where the highest compressive strength was observed at a 30% gold tailings proportion. But at 40% replacement of sand with gold tailings, the compressive strength of the cement mortar decreases. This is specifically observed from the study that the optimum replacement of sand is 30%. For concrete applications, other researchers also observed the same trend: the decrease in compressive and flexural strength decreases with the increase in tailings proportion [12].

The decrease in compressive strength of the cement-based mortar with T'Boli gold tailings could be attributed to the lack of component coalescence [24] because of the insufficient CaO to SiO_2 ratio. Increasing the proportion of gold tailings decreases the proportion of the binder, which is cement. Cement contributes mainly CaO components to the cement mortar matrix. Decreasing its proportion means a lack of CaO to bind with excess SiO_2 in the formation of calcium silica hydrates, dicalcium silicate, and tricalcium silicate, which are considered to be the main strength contributors of concrete and cement mortar. The compressive strength of the cement mortar, both control and with T'Boli gold tailings, increases as curing days increase [13]. This is because prolonged curing days provide more time for cement hydration [7, 28].

The Department of Transport in the UK has required at least 4.5 and 15 MPa compressive strengths of cement mortar to qualify as a component in the sub-base and base of roadways [29]. Sub-base and base layers for road construction are one of the most important layers in road structure considered in highway engineering. Sub-base is usually composed of granular and cement materials; however, cement mortar is widely utilized for the construction of sub-base in roadways [30]. With the current compressive strength of the cement-based mortar with T'Boli gold tailings, this material possesses great potential as a component for the sub-base layer in roadway construction.

4.0 CONCLUSION

This study investigated the viability of T'Boli gold tailings as alternative to sand (fine aggregates) in developing cement mortar. The main conclusions are as follows:

- i T'Boli gold tailings' particle size was found to be 58.2% passing an 80-mesh sieve (180 µm), which is much finer than ordinary sand that is typically used in cement mortar production.
- ii The X-ray fluorescence (XRF) analysis shows a great abundance of SiO_2, Al_2O_3, Fe_2O_3, K_2O, and CaO minerals, which indicates that T'Boli gold tailings have a pozzolanic nature.
- iii The scanning electron microscopy with energy dispersive X-ray (SEM-EDX) spectroscopy revealed that T'Boli gold tailings have irregular-shaped flakes and granular particles with a relatively large abundance of oxygen, silicon, and aluminum elements.
- iv The highest compressive strength observed from cement-based mortar with T'Boli tailings is 9.11 MPa, which was formulated with 80% by weight of T'Boli gold tailings and water cured for 14 days.
- v The compressive strength test shows that as the proportion of gold tailings in cement mortar increases, there is a decrease in its compressive strength. This is due to the lack of component coalescence because of an inadequate SiO_2 and CaO ratio, inhibiting the formation of calcium silica hydrates, dicalcium silicate, and tricalcium silicate.
- vi Lastly, the compressive strength of cement-based mortar with T'Boli gold tailings increases as the curing period increases.

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