



Research Article

EVALUATION OF GOLD MINE TAILINGS IN CEMENT MORTAR: INVESTIGATION OF THE EFFECTS OF CHEMICAL ADMIXTURES

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ABSTRACT

This study presents an experimental investigation on the influences of gold mine tailings as a substitute for cement. Various ratios of air-entraining and superplasticizer admixtures were used in the determined composition of gold mine tailing-containing mortars. Several analytical variables, including setting time, water absorption, and strength tests, were studied to establish the relationship between these added materials and the properties of cement mortars. In addition, the compositions of 28-day-cured samples were identified by X-ray diffraction (XRD) analysis. The results showed that when the water /binder (w/b) ratio is kept constant at 0.5, the replacement of cement by 10% gold mine tailing showed mechanical properties similar to those of the reference sample. The mortars prepared with 0.5% superplasticizer admixture reached the highest compressive and flexural strength values of 64.16 MPa and 8.99 MPa, respectively. When flowability and water absorption properties are improved by the air-entraining admixture addition, the compressive and flexural strength decrease.

Keywords: Cement mortar, chemical admixture, gold mine tailing, industrial waste.

1. INTRODUCTION

Currently, with the population increasing and excessive use of available resources, reduction and/or recycling of wastes and evaluation of waste as a potential raw material source have gained importance. The use of industrial wastes in the construction industry ensures a decrease in energy costs and environmental pollution resulting from less cement use, which demands high energy consumption and CO₂ emission [1, 2].

As a fine-grained sludge, gold mine tailings occur as waste of the gold extraction process and their disposal is a major environmental problem [3, 4]. Each year, approximately 278 000 tons of tailings are produced during gold extraction process in the Ovacık Gold Mine in Turkey [5, 6]. After treatment of ores, the Inco SO₂/Air process is used to control cyanide effluents, and the tailings are stored in a pond with a surface area of 16000 m², a 30 m embankment height, and a storage capacity of 1.6 million m³ [7]. In terms of both economic and environmental effects,

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instead of storing in these ponds, reusing the tailings in various industrial applications gains importance.

Industrial wastes or by-products like fly ash, silica fume, bottom ash, and slag have been evaluated in the construction industry for many years. Besides environmental benefits, the chemical composition of these materials helps to reduce costs in construction technologies and improves properties of construction materials. Conversely, because of the mineral contents of complex gold deposits, (the variety of rocks, rare deposits), gold mine tailings can be used directly as an additive material [8-12]. However, studies of the utilization of gold mine tailings as supplemental materials in the composition of cement composites are limited. In one study [13], it was reported that the usage of gold mine tailings as a fine aggregate increased the compressive strength of cement mortar. In the study conducted by Mahmood and Mulligan [8], the influence of six different types of tailings from several mines in Canada (copper, gold, and others) on the unpaved road base construction was investigated. The results showed that all tailings met the minimum strength limit required for filling underground stopes. Vignesh et al. [14] established that gold mine tailings can be used as a natural sand in the production of masonry mortars. Ramesh et al. [15] used the gold tailings with lime to improve properties of red soil. Also, the possibility of using of gold mine tailings for making bricks was researched by Roy et al. [5] it was reported that gold tailings were useful as an additive material in this process. Fall et al. [16] investigated the effect of gold tailings, fly ash, and blast furnace slag on compressive strength of cemented paste backfill. Based on the compressive strength of the 28-day-cured samples, the combination of gold tailings with blast furnace slag showed better mechanical performance than a gold tailings and fly ash mix. Kunt et al. [6] and Celik et al. [17] evaluated gold tailings as an additive material in cement mortar production; they found that tailings are workable as the mortar aggregate.

In this study, the aim was to evaluate gold mine tailings instead of cement in the mortars. For this purpose, effects of the different ratio of gold mine tailings, which were obtained from the Bergama gold mine site (Izmir, Turkey), and water/binder ratio on properties of cement mortar were investigated. Additionally, air-entraining and superplasticizer admixtures were used to improve properties of mortars modified with gold mine tailings. The fresh and hardened mortars were tested for setting time, consistency, compressive and flexure strength. Also, X-ray diffractometer (XRD) was used for crystallographic analysis of the mortars.

2. MATERIALS AND METHODS

2.1. Materials

Ordinary Portland cement CEM I 42.5R, compliant with Turkish Standard TS EN 196 [18], was used to prepare cement mortars; it was obtained from the Akçansa Cement Factory (Istanbul, Turkey). The gold mine tailings were supplied by the Bergama Gold Mine Treatment Plant (Izmir, Turkey) and maintained at 105°C to remove the water content. The dried tailings were ground with Retsch RM 100 (Retsch GmbH & Co KG, Haan, Germany) and sieved with a Fritsch analysette 3 Spartan pulverisette 0 vibratory sieve shaker (Fritsch, Idar-Oberstein, Germany). Rilem Cembureau sand, which was used as a fine aggregate, was supplied by the Limak Trakya Cement Plant (Kirkclareli, Turkey). The Blaine surface area of cement and tailings were 3666 cm²/g and 2888 cm²/g, respectively. The chemical compositions as determined by X-ray fluorescence (XRF) (Philips PANalytical Minipal 4) is shown in Table 1.

Polycarboxylic ether-based superplasticizer (MasterGlenium®51) and an air-entraining admixture (MicroAir 200) were obtained from BASF Construction Chemicals (BASF Turkey). The technical properties of chemical admixtures are shown in Table 2.

Table 1. Chemical composition of cement, gold mine tailing and sand

| Composition (%) | Cement | Gold mine tailing | Sand |
|--------------------------------|--------|-------------------|------|
| SiO ₂ | 13.00 | 89.25 | 90.8 |
| Al ₂ O ₃ | 2.00 | 6.10 | 5.70 |
| Fe ₂ O ₃ | 4.70 | 0.84 | 0.86 |
| CaO | 71.8 | - | 0.41 |
| SO ₃ | 2.90 | 0.09 | - |
| MgO | 4.00 | 0.29 | - |
| TiO ₂ | - | - | 0.87 |
| K ₂ O | 1.20 | 1.24 | 2.90 |
| Loss of ignition | 2.20 | 2.53 | 2.50 |

Table 2. Technical properties of chemical admixtures

| Chemical admixture | MasterGlenium®51 | MicroAir 200 |
|----------------------|----------------------|-------------------------------|
| Composition | Polycarboxylic ether | Oil alcohol and ammonium salt |
| Density (kg/L) | 1.082-1.142 | 0.98-1.02 |
| Alkaline Content (%) | < 3 | < 10 |
| Chloride Content (%) | < 0.1 | < 0.1 |

2.1. Mixture proportions and sample preparation

To investigate the suitability of gold mine tailing as a substitute in cement mortar, different ratios (between 5% and 25% by weight) of tailings were used with three different water binder ratios: 0.50, 0.45, and 0.40. The optimum w/b ratio and gold mine tailing dosage were determined based on mechanical properties of mortars. In the following step of experimental study, superplasticizer and air-entraining admixtures were used in the mixtures at the ratios of 0.1% to 0.2% and 0.4% to 0.5% of binder materials by weight, respectively. From preliminary studies, the optimal gold mine tailing was determined to be 10% for the chemical admixture-containing mortars. Also, the w/b ratio was chosen as 0.40 because the used chemical admixtures decrease the water demand of the mortars. For each w/b ratio, the reference mixtures consisted of only Portland cement, sand (1350 g), and water without any chemical admixtures. All samples, which were prepared according to the EN 196-1 standard [18], were demoulded after 24 h and cured in the water at 20°C to specified ages of testing (3, 7, and 28 days). Table 3 presents the detailed mix proportions of the prepared cement mortars and Figure 1 shows the experimental steps.

Table 3. Mixing proportions of mortar

| Mix | Cement (%) | BG (%) | Sand (g) | Air-entraining admixture (%) | Superplasticizer admixture (%) | w/b ratio |
|------------|------------|--------|----------|------------------------------|--------------------------------|-----------|
| RF-0.50-00 | 100 | 0 | 1350 | - | - | 0.50 |
| BG-0.50-05 | 95 | 5 | 1350 | - | - | 0.50 |
| BG-0.50-10 | 90 | 10 | 1350 | - | - | 0.50 |
| BG-0.50-15 | 85 | 15 | 1350 | - | - | 0.50 |
| BG-0.50-20 | 80 | 20 | 1350 | - | - | 0.50 |
| BG-0.50-25 | 75 | 25 | 1350 | - | - | 0.50 |
| RF-0.45-00 | 100 | 0 | 1350 | - | - | 0.45 |
| BG-0.45-05 | 95 | 5 | 1350 | - | - | 0.45 |
| BG-0.45-10 | 90 | 10 | 1350 | - | - | 0.45 |
| BG-0.45-15 | 85 | 15 | 1350 | - | - | 0.45 |
| BG-0.45-20 | 80 | 20 | 1350 | - | - | 0.45 |
| BG-0.45-25 | 75 | 25 | 1350 | - | - | 0.45 |
| RF-0.40-00 | 100 | 0 | 1350 | - | - | 0.40 |
| BG-0.40-05 | 95 | 5 | 1350 | - | - | 0.40 |
| BG-0.40-10 | 90 | 10 | 1350 | - | - | 0.40 |
| BG-0.40-15 | 85 | 15 | 1350 | - | - | 0.40 |
| BG-0.40-20 | 80 | 20 | 1350 | - | - | 0.40 |
| BG-0.40-25 | 75 | 25 | 1350 | - | - | 0.40 |
| BG-AE-0.10 | 90 | 10 | 1350 | 0.10 | - | 0.40 |
| BG-AE-0.15 | 90 | 10 | 1350 | 0.15 | - | 0.40 |
| BG-AE-0.20 | 90 | 10 | 1350 | 0.20 | - | 0.40 |
| BG-SP-0.40 | 90 | 10 | 1350 | - | 0.40 | 0.40 |
| BG-SP-0.45 | 90 | 10 | 1350 | - | 0.45 | 0.40 |
| BG-SP-0.50 | 90 | 10 | 1350 | - | 0.50 | 0.40 |

*BG: Gold mine tailing, AE: Air entraining admixture, SP: Superplasticizer

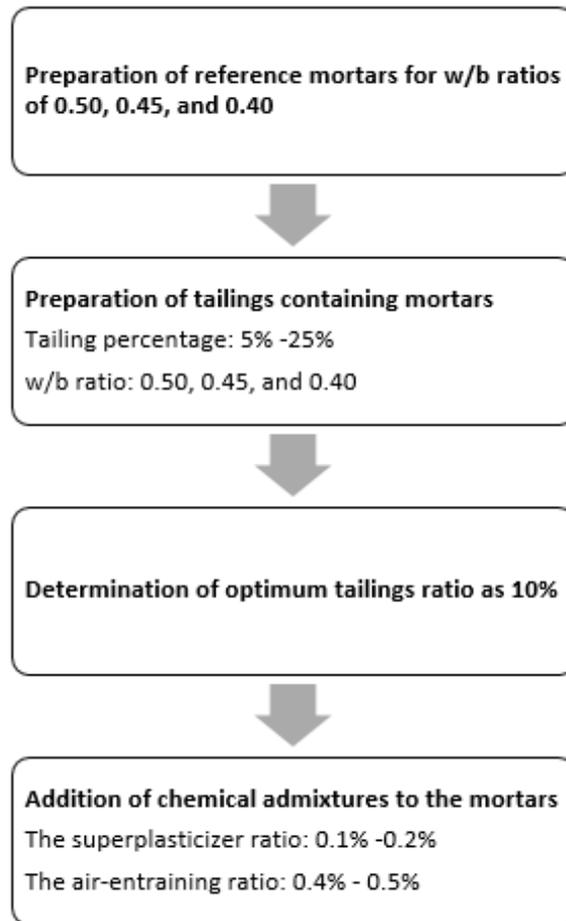


Figure 1. The experimental steps

2.3. Testing specimens

The consistency of fresh mortar was determined by the flow table test as specified by EN 1015-3 after mixing [19]. The determination of initial and final setting times of prepared mortars were performed by a Vicat apparatus based on penetration of Vicat needles, as based on TS EN 480-2 [20].

To observe the influence of various experimental parameters on different age strengths of mortars, the compressive and flexural strength of hardened mortars were measured by a UTEST brand automatic cement compression- and flexure-testing machine. Initially, the flexural strength test was applied to specimens and the obtained two halves were used for compressive strength. Specimens were subjected to a water absorption test after 24 hours of immersion in the curing water. For this experiment, initial (M_i) and final mass (M_f) of specimens were measured after drying their surface with a towel; the percentage of absorbed water (A) was calculated from the mass difference (Equation 1).

$$A = (M_f - M_i) / M_i \times 100 \quad (1)$$

PANalytical Xpert Pro (PANalytical B.V., Almelo, The Netherlands) X-ray diffractometer was used to identify crystalline phases of cement mortars in the pattern range of 5° to 90°, at the scanning rate of 0.006°/s and Cu-K α radiation at 45 kV and 40 mA.

3. RESULTS AND DISCUSSION

3.1 Fresh properties of mortars

The results of flow table test for different w/b ratios and chemical additive contents are shown in Figure 2 and Figure 3, respectively. When different w/b ratios were compared, the flow diameters of mortars were highest for w/b ratio of 0.50. Additionally, for the w/b ratios of 0.50 and 0.40, the increasing dosages of gold mine tailings enhanced the workability.

This observation can be explained by surface area differences of the cement and the gold mine tailings: As the cement is finer than the gold mine tailings, the water demand of mortars decreases with decreasing cement content.

After determining the optimal gold mine tailing amount as 10% of binder and a w/b ratio as 0.40, the previously mentioned chemical additive dosages were added to the binder. Figure 3 shows that the amount of entrained air modified the rheology of the cement mortar. The dosage of air entraining admixture affected the flow diameter proportionately. However, unexpectedly, the varied dosages of superplasticizer could not create a significant difference in the rheological properties. The effects of superplasticizer on the flowability depends on adsorption of superplasticizer by the cement particles. When the adsorbed superplasticizer amount reaches its saturation point, the fluidity of the cement composites is minimally affected by an increasing dosage of superplasticizer [21].

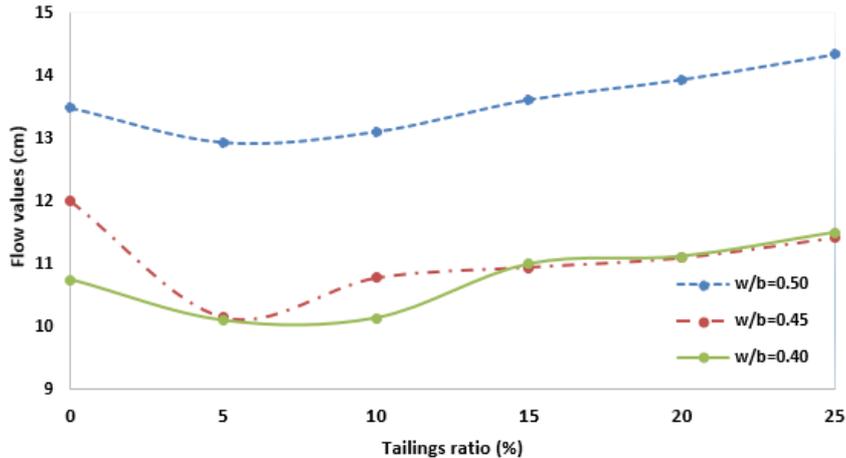


Figure 2. The flow diameters of fresh mortars for different w/b ratios

The initial set was seen to occur at 175 min, 165 min, and 160 min; the final set was observed at 330 min, 300 min, and 270 min for RF-0.50-00, RF-0.45-00, and RF-0.40-00 samples, respectively. Some studies have shown that replacement of mine tailings has a negative influence on the setting time because of the heavy metal content of tailings [22, 23]. Although the heavy metal content of gold mine tailing used is negligible, with a few exceptions, the replacement of gold mine tailings delayed the setting progress, especially for a w/b ratio of 0.50. This phenomenon can be explained by the lower activity of tailings. However, addition both air-

entraining and superplasticizer admixtures, generally accelerated the initial and final setting in comparison with BG-0.40-10. For the chemical admixture-containing mortars, BG-0.40-10 was chosen as the control mixture because the same w/b and tailings ratios were used to prepare the chemical additive-containing mixtures. Conversely, when a 0.20% air-entraining admixture was added to mortar, the initial setting time was delayed up to 5.71% of the control mortar.

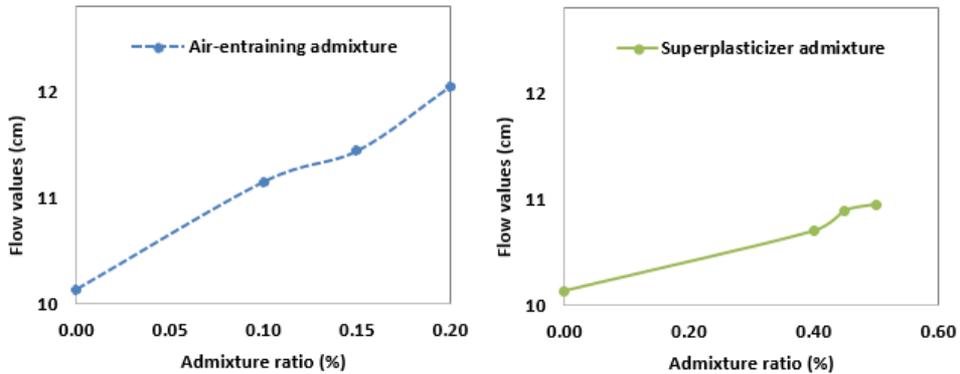


Figure 3. The flow diameters of fresh mortars for different chemical admixtures

Table 4. The relative change in initial and final setting time

| Mix | Initial setting time change (%) | Final setting time change (%) |
|------------|---------------------------------|-------------------------------|
| RF-0.50-00 | 0.00 | 0.00 |
| BG-0.50-05 | 31.43 | -13.64 |
| BG-0.50-10 | 25.71 | 9.09 |
| BG-0.50-15 | 20.00 | 4.55 |
| BG-0.50-20 | 8.57 | 0.00 |
| BG-0.50-25 | 5.71 | -4.55 |
| RF-0.45-00 | 0.00 | 0.00 |
| BG-0.45-05 | 9.09 | 5.00 |
| BG-0.45-10 | 3.03 | 0.00 |
| BG-0.45-15 | 6.06 | 0.00 |
| BG-0.45-20 | 3.03 | -5.00 |
| BG-0.45-25 | 0.00 | -10.00 |
| RF-0.40-00 | 0.00 | 0.00 |
| BG-0.40-05 | 12.50 | 16.67 |
| BG-0.40-10 | 9.38 | 11.11 |
| BG-0.40-15 | 6.25 | 5.56 |
| BG-0.40-20 | -12.50 | 0.00 |
| BG-0.40-25 | -15.63 | -11.11 |
| BG-AE-0.10 | -17.14 | -10.00 |
| BG-AE-0.15 | -5.71 | -5.00 |
| BG-AE-0.20 | 5.71 | 0.00 |
| BG-SP-0.40 | -11.43 | -25.00 |
| BG-SP-0.45 | -8.57 | -20.00 |
| BG-SP-0.50 | -2.86 | -15.00 |

The relative change in initial and final setting time of prepared cement mortars is given in Table 4. The delay of setting time by increasing the amount of air-entraining admixture result from the adsorption of surfactant molecules by the binder: water molecules are blocked, and the topochemical reaction between binder and water is inhibited. Hence, the hydration reactions are retarded [24].

3.2. Water absorption of mortars

Water absorption is one of the most important and relevant parameters for the durability of cement composites. The solid and pore phases in the mortars create a matrix that provides a flow of water into the structure and affects the properties of mortars [24-26].

Figure 4 shows the water absorption percentage of 1 day cured mortars with and without chemical admixtures: it shows that mortars prepared with a 0.40 w/b ratio exhibited higher water absorption percentages, especially for the lower amount of added gold mine tailing. However, using w/b ratios of 0.45 and 0.50 significantly reduced the amount of absorbed water, which showed that less porous structures were obtained by higher w/b ratios. In general, the increasing amount of tailing decreased the absorbed water; that can be explained by the fineness of the cement particles. As cement is finer than tailings, when a given amount of cement is replaced by tailing, the water content of mortar is at a level sufficient for further hydration.

The addition of the air-entraining and superplasticizer admixtures reduced the absorbed water in comparison with the reference mortar (BG-0.40-10). When the water absorption percentage was 1.49% for BG-0.40-10 sample, the water absorption values were changed from 0.63% to 0.52%, and 0.79% to 0.75%, for air-entraining and superplasticizer admixtures, respectively. Previous studies showed that using adequate amounts of air-entraining or superplasticizer admixtures decreased the amount of absorbed water [27-29].

The highest compressive strength value for a curing time of 28 days was measured as 51.19 MPa for the BG-0.50-10 sample, when the compressive strength of reference sample (RF-0.50-00) was 54.67 MPa. However, the hardening of cement composites was within acceptable limits up to 15% of the replacement ratios of tailings. Based on compressive strength results, the w/b ratio of 0.40 was not suitable because of low strength values. Although Abrams' water/cement ratio law reveals the inverse relation between compressive strength and w/b ratios, the results obtained in the present study were not matched to this approach [30] for a w/b ratio 0.40, a less integrated matrix was obtained because of lack of water for further hydration.

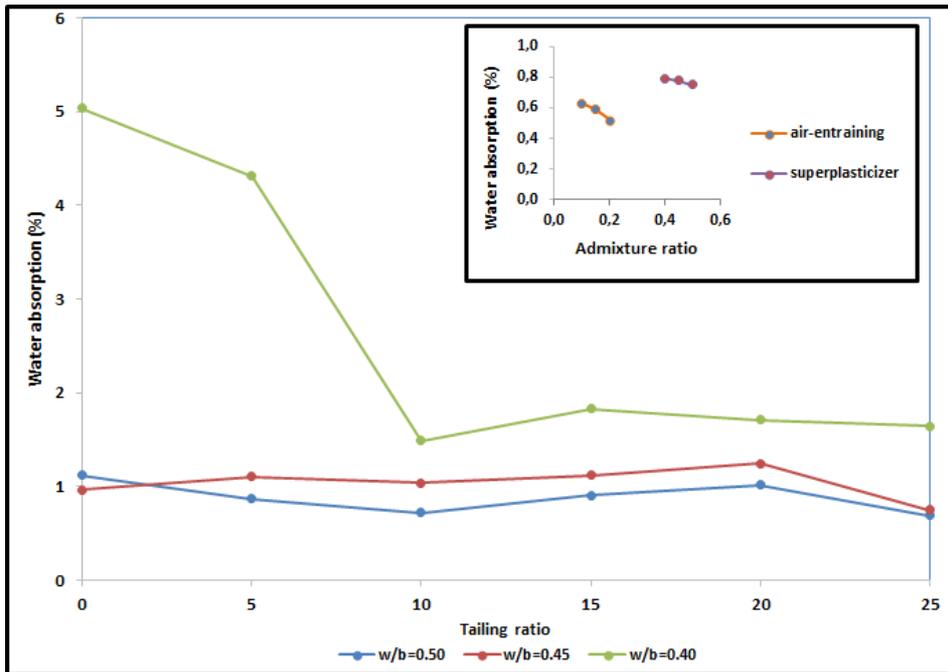


Figure 4. Water absorption percentages of mortars with and without chemical additive

3.3. Compressive and flexural strength of mortars

The average compressive strength of mortars for cement composites are presented in Figure 5. Resulting from the hydration process, the compressive strength of mortars increased during the curing period. For the w/b ratios of 0.50 and 0.45, for all test days, addition of increasing amounts of gold mine tailing affected compressive strength adversely.

The air-entraining admixture increases the volume of air in the mortar matrix to enhance the workability, and as a result, the compressive strength shows a decrease [31]. Figure 5 shows that the addition of the air-entraining admixture caused a significant decrease in compressive strength for all w/b ratios. The compressive strength of 28-days-cured samples ranged between 9.05 MPa and 26.36 MPa.

As expected, the contribution of superplasticizer in the mortar with increasing ratios caused an increase in compressive strength. The compressive strength of the mortars with 0.40%, 0.45%, and 0.50% superplasticizer after 28 days of hardening was 50.41 MPa, 52.94 MPa, and 64.16 MPa, respectively. The results showed that using 0.50% superplasticizer in gold mine tailing-containing mortars can improve the compressive strength by 25.34% of the BG-0.50-10 sample, which had the highest compressive strength and contained no additional admixture. In a previous study [13], the highest compressive strength value was 36.8 MPa for 28 days curing when gold mine tailings were used as fine aggregate. The results revealed that the introduction of superplasticizer admixture induced a more compact matrix because the adsorption of superplasticizer increased the dispersion on the cement grains and water interface.

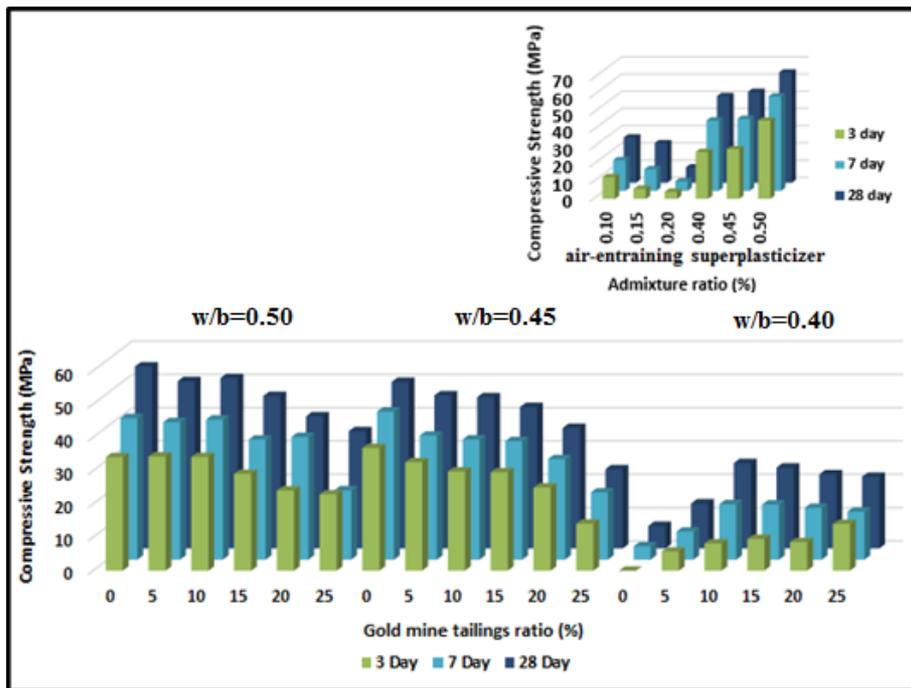


Figure 5. Compressive strength of mortars

Figure 6 shows the flexural strengths of the prepared cement mortars. The mortars prepared with various amounts of gold mine tailing and w/b ratios had lower flexural strength than the reference samples for all w/b ratios. The increasing amounts of gold mine tailings caused a decrease in the flexural strength for the w/b ratios of 0.50 and 0.45. When the w/b ratio was reduced to 0.40, due to the lack of component coalescence, a sharp decrease was observed for all tailing amounts. Differently, mortars with w/b ratios of 0.50 and 0.45 showed higher flexural strength by increasing tailing content for w/b ratio of 0.40. The decrease in the flexural strength by increasing tailing content for w/b ratios of 0.50 and 0.45 can be explained by lower pozzolanic activity of tailings. Furthermore, when flexural strength is a function of the porosity of the material, using tailings at increasing ratios can result in less improvement in flexural strength due to its Blaine surface area. Nevertheless, the flexural strength development showed a different trend for w/b ratio of 0.4. The less amount of cement used in the content of cement mortar leads to a lower water demand for hydration reactions. For a w/b ratio of 0.4, because of cement fineness, increasing tailing content induced an increase of flexural strength, except for a tailing addition at 20%. Like compressive strength, addition of air-entraining admixture had reverse effects on the flexural strength. The highest flexural strength was obtained for an air-entraining admixture ratio of 0.10% at 2.52 MPa, 3.79 MPa, and 4.91 MPa for the curing days of 3, 7, and 28, respectively. At the end of the 28 days of curing, the flexural strength for superplasticizer ratios of 0.40%, 0.45%, and 0.50% was 7.40 MPa, 7.86 MPa, and 8.99 MPa, respectively. These results showed that use of 0.50% superplasticizer with gold mine tailings can increase the flexural strength by 20.03% of BG-0.50-10 sample.

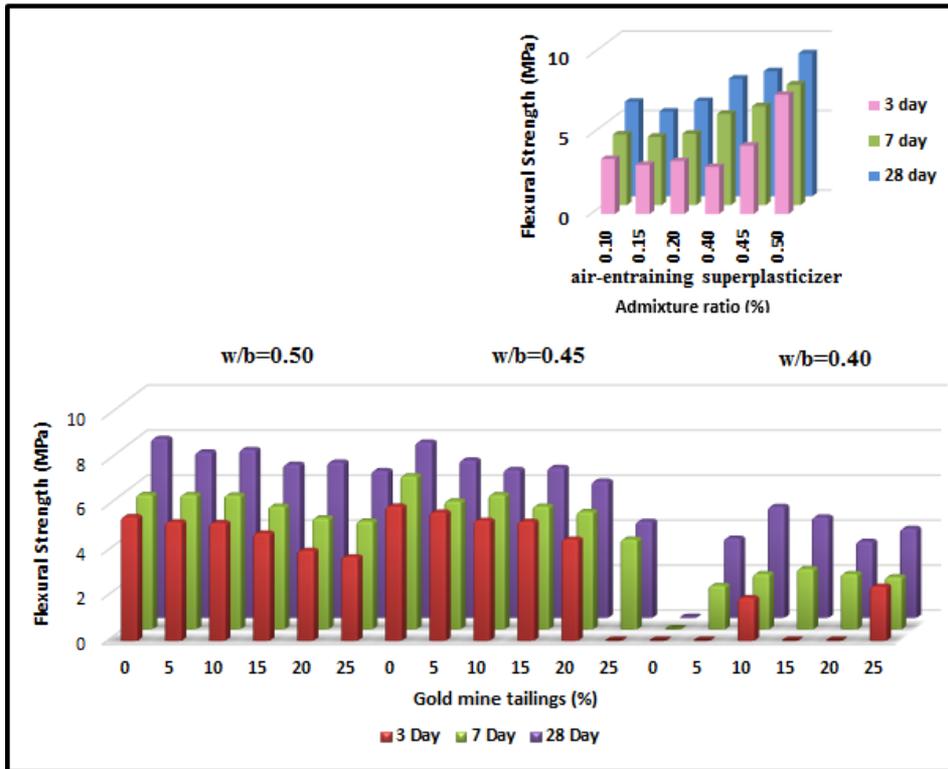


Figure 6. Flexural strength of mortars

3.4. XRD Analysis

XRD patterns of 28 day-cured samples, which showed the highest compressive strength for each set, are depicted in Figure 7. According to XRD results, portlandite ($\text{Ca}(\text{OH})_2$), quartz (SiO_2), calcium alumina silicate (C-A-S), and calcium silicate hydrate (C-S-H) were identified as the crystalline components of cement mortars. The hydration products had a less crystalline structure, which caused small peaks. In the XRD patterns, the major peaks were related to quartz that came from sand. In XRD analysis, the progress in the hydration can be followed based on the intensity change of portlandite peaks. The consumption of portlandite during hydration (Equations (2-4)) can be approximated by the intensity decrease of main diffraction peaks of CH crystals (at $2\theta = 18^\circ$ and 34°) [32].



For samples BG-0.4-10, BG-0.45-10, and BG-EA-0.1, the intensity of portlandite peaks was higher than that of BG-0.5-10 and BG-SP-0.5. This phenomenon is in agreement with compressive and flexural strength test results. With 28-day curing, the flexural strength of BG-0.4-10, BG-0.45-10, and BG-EA-0.1 specimens were 4.96 MPa, 6.60 MPa, and 4.91 MPa, respectively. BG-0.5-10 and BG-SP-0.5 had flexural strengths of 7.49 MPa and 8.99 MPa, respectively. The peak intensities of calcium silicate hydrate crystals are higher than other

samples for BG-0.5-10 and BG-SP-0.5. The results indicate that using a w/b ratio of 0.5 or addition of a superplasticizer admixture can improve the strength properties by consuming more portlandite crystals at the interface and allowing further hydration reactions.

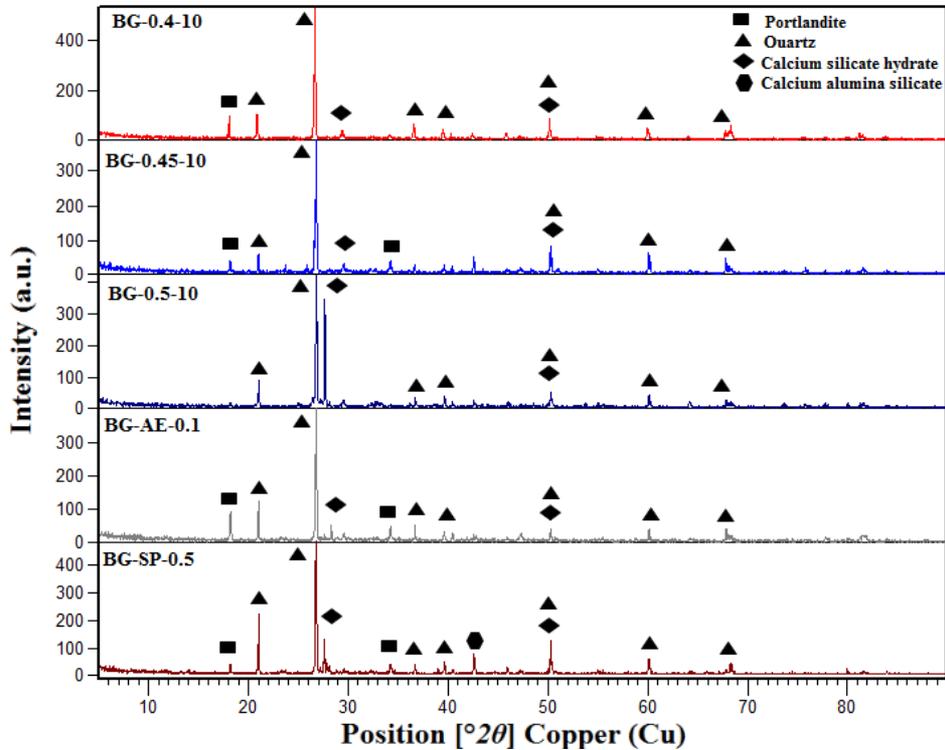


Figure 7. XRD patterns of mortars

4. CONCLUSION

The present study was focused on recycling gold mine tailings in the content of cement mortar as a mineral additive and investigated the effects of air-entraining and superplasticizer admixtures. The obtained results showed that:

1. The replacement of gold mine tailings shortened the initial and final setting time for w/b ratio of 0.5. Also, the chemical admixtures improved setting progress for admixture ratios.
2. The increasing w/b ratio increased the flowability of the mortars as expected. When the addition of air-entraining developed the workability by increasing ratios, the superplasticizer addition did not have a significant effect on the flowability.
3. The gold mine tailings-containing mortars prepared with w/b ratios of 0.45 and 0.50 had lower water absorption than reference samples, which was a result of higher hydration reactions. Superplasticizer admixture decreased the absorbed water amount up to 49.7% of reference sample when air-entraining containing mortars absorbed 65.1% less water than reference.
4. Despite being contrary to Abrams' w/b law, compressive and flexural strength of the gold mine tailing-containing mortars reached higher values for the w/b ratio of 0.50. Results indicated that increasing amount of gold mine tailings as a replacement decreases mechanical endurance, which could result from the decrease in amount of cement.

5. The mortars including a 0.5% superplasticizer admixture reached the highest compressive and flexural strength values at 64.16 MPa and 8.99 MPa, respectively. However, the combined use of gold mine tailings and air-entraining admixture decreased the compressive and flexural strength values.

6. The addition of superplasticizer ensured more compact, impermeable and enduring mortars than other samples. Moreover, the usage of mine tailings with optimum amount of superplasticizer can be effective to reduce environmental effects besides its positive economic contribution.

Acknowledgments

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