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A mechanical and environmental approach on the partial substitution for aggregate in mortar using various metal mine tailings

Introduction

The raw material requirements of many industries are met by mining activities. As a result of these mining activities, a significant amount of tailing is formed. If these tailings are not managed correctly, they can cause critical environmental problems. For instance, when sulfur-bearing minerals are exposed to atmospheric oxygen and water, sulfuric acid is formed. Sulfuric acid dissolves heavy metals such as As, Cd, Cr, Cu, Pb, Ni, Zn, etc.,

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and high concentrations of these ions cause groundwater and soil pollution, which creates considerable negative consequences on living organisms (Ali and Khan 2018; Iatan 2021; Kapoor and Singh 2021; Zhang et al. 2023). Therefore, the correct management of these tailings is a necessity. For this reason, the tailings must be stored and removed under certain safety conditions, which increases the overall costs of the mining activities. Cacciuttolo and Atencio (2023) reported that the cost per ton of conventional tailings transported from the beneficiation plant to the tailings storage facility was 1 USD, while this cost was 2 USD for thickened tailings, 3 USD for paste tailings, and 4 USD for filtered tailings. On the other hand, the cost of in-pit disposal of mine tailings is 2.5 USD per ton. In this context, the use of these tailings as a substitute material in an adequate process stands out as a good alternative as it reduces the amount of tailings and hence mine production costs, as well as creating additional income. Reusing mine tailings tends to preserve natural resources, reduce environmental impact, and contribute to economic growth by 9% (Cheng et al. 2016; Ikopbo and Boado 2024).

One of the industries where mine tailings can be utilized is concrete production. Concrete is probably the most important building material that forms the basis of many structures, including buildings, roads, bridges, and dams. It is estimated that more than 10^9 m^3 of concrete is produced worldwide annually. This amount means approximately 3 tons of concrete per person annually, which makes concrete the most produced and consumed material following water (Gagg 2014).

Approximately 70% of the volume of concrete consists of aggregate (crushed stone, gravel, and sand), in addition to 10% cement and 20% water. Nowadays, since the supply of washed-screened sand and gravel is quite limited, screened crushed stones are mostly used as aggregate in concrete production.

In this context, in the crushed stones used as aggregates, the size fraction below 0.063 mm that can exist as dispersed, agglomerated, or even attached to large aggregate particles is of great importance in the strength of the concrete. As the fine aggregate amount in concrete increases, the water amount required to produce concrete with a constant consistency increases. Furthermore, the increase in the amount of water increases the shrinkage and porosity of the concrete, which decreases its strength. Meanwhile, mine tailings, especially those with low clay content, can be used as a substitution for fine aggregates in concrete as an inert filler with the advantage of accelerating hydration reactions by creating nucleation sites for hydrates in cement, and hence improving concrete compressive strength and providing a heterogeneous nucleation effect (Ye et al. 2007; Sigvardsen et al. 2018). In the EN 12620 standard, which specifies the properties and performance requirements of aggregates used in concrete production, it is stated that the fraction under 0.063 mm will not affect the strength of the concrete negatively if its amount is less than 3% (EN 12620). This standard also states that if the ratio of this fraction is over 3%, methylene blue (EN 933-9) or sand equivalence (EN 933-8) tests should be performed to determine the clay content. The results of these tests should meet the limit values of the mortar strength tests (Shilstone 1990; Ye et al. 2007; Sigvardsen et al. 2018; EN 12620; EN 933-9; EN 933-8; Okonkwo and Arinze 2018; Amini et al. 2019).

Using mine tailings as a partial substitution for the aggregates can reduce the raw material requirement in concrete production. In addition, mine tailings have undergone size reduction processes due to mining and mineral processing operations and generally exist in particle sizes below 0.500 mm. Therefore, size reduction activities applied for aggregate production are not required for mine tailings. Therefore, the overall energy consumption and costs in concrete production will be decreased in the case of using these tailings. Furthermore, the carbon footprint, which is a constraint for sustainable concrete production, will also be reduced (Flower and Sanjayan 2007; Adiguzel 2012; Ferreira et al. 2015; Bascetin et al. 2016; Bascetin et al. 2017).

In recent years, the effectiveness of the use of mine tailings, especially in ceramic, glass, and high-performance concrete products, has been investigated, and promising results have been obtained (Choi et al. 2009; Lottermoser 2010; Zhao et al. 2014; Zhu et al. 2015; Adiguzel et al. 2022). In previous studies, iron, lead-zinc, copper, molybdenum, gold, coal, magnesite, boron, and marble mine tailings were used as aggregate or cement substitutes. In these studies, mortar samples were produced by increasing the tailing substitution ratio up to 50%, and uniaxial compression strength (UCS) tests were carried out to determine the effect of the tailing type and substitution ratio on the strength of the mortar samples. As a result of these studies, it is seen that the mechanical properties of the concretes vary significantly with various parameters, including the tailing type and substitution ratio (Aliabdo et al. 2014; Yunhong et al. 2016; Pavez et al. 2016; Carrasco et al. 2017; Liang et al. 2017; Ince 2019; Esmaeili et al. 2020; Gao et al. 2020; Bagger et al. 2021; Shanmugasundaram and Shanmugam 2021; Zhao et al. 2021). Therefore, it is crucial to reveal the physical and chemical properties of the tailings in terms of mechanical and environmental aspects. However, since the physical and chemical properties of mine tailings vary in a wide range, a standard has not yet been established for their use in concrete production, contrary to various pozzolanic materials such as fly ash, slag, and silica fume (EN 450-1; Pinheiro et al. 2024). Accordingly, the use of various mine tailings in concrete production is still an interesting field of research (Martins et al. 2021; Chen et al. 2022; Méndez et al. 2022; Ikotun et al. 2024).

Therefore, in this study, the usability of four different metallic mine tailings of gold, lead-zinc, copper, and iron, with different chemical and physical properties, as aggregate substitution material in concrete was investigated in terms of mechanical and environmental aspects. Following this purpose, uniaxial compressive strength (UCS) tests were carried out at various substitution ratios for each tailing as a function of curing time. Thus, the maximum possible substitution ratio for each tailing was revealed in terms of the strength of the concrete. In addition, the pH level of the tailings before and after the concrete production was measured as a function of time. Therefore, the risk of dissolved heavy metal mobilization that may occur at low pH levels and the environmental pollution potential were investigated in terms of acid mine drainage (AMD) for the tailings and tailing-substituted mortar samples.

1. Materials

The mine tailing samples used in this study were supplied from the discharge points of the mineral processing plants of gold (İlic, Erzincan), lead-zinc (Balya, Balıkesir), copper (Kure, Kastamonu), and iron (Divrigi, Sivas) mines in Türkiye, and coded as AuT, LZT, CuT, and FeT for the experiments, respectively.

As is known, when sulfurous minerals such as pyrite come into contact with atmospheric oxygen and water, sulfuric acid is formed. The sulfuric acid dissolves heavy metals in the medium and forms an acidic solution containing high concentrations of heavy metals. This solution, which is called acid mine drainage (AMD), seeps into the soil, pollutes surface and groundwater, and has a significant impact on the environment (Moosberg-Bustnes et al. 2004; Zhao et al. 2014). To reduce the AMD risk, cement binders can be added to the mine tailings (Bascetin and Tuylu 2018; Tuylu et al. 2019; Iatan 2021).

In addition, heavy metals are considered among the most important water pollutants due to the toxic and mutagenic effects they cause by accumulating in various body organs even at low concentrations (Ghosh et al. 2012; Wongrod et al. 2018; Reta and Mekonen 2023). For this reason, the World Health Organization (WHO) has determined an upper limit for each heavy metal in terms of human health (WHO 2006). In this regard, the heavy metal contents of the tailings used in this study were determined by inductively coupled plasma – mass spectrometry (ICP-MS) analysis. The results are given in Table 1, along with the limit values for human health determined by the WHO.

Table 1. Heavy metal contents of the tailings and WHO limits

Tabela 1. Zawartość metali ciężkich w odpadach poflotacyjnych i limity WHO

Heavy metal	AuT (ppm)	LZT (ppm)	CuT (ppm)	FeT (ppm)	Upper limit (by WHO 2016)
Ag	8.7	3.2	2.3	0.4	0.1
As	3,139.9	417.1	189.1	57.1	0.01
Cd	1.2	3.6	1.1	0.1	0.05
Cr	52.8	49.7	232.3	108.4	0.003
Cu	1,702.6	155.3	1,614.2	81.1	0.02
Mn	5,492.2	2,099.1	1,090.4	373.2	0.02
Pb	39.2	667.1	62.8	29.6	0.01
Se	2.3	2.3	12.8	0.5	0.02
Zn	261.5	841.4	915.1	46.7	3.00

As can be seen in Table 1, the heavy metal content of the tailings was quite above the WHO's upper limit values. Therefore, releasing these materials into nature in an uncontrolled way is very risky in terms of human health and the environment. On the other hand, storing these tailings in durable materials such as concrete prevents the release of heavy metals into nature and provides various environmental advantages (Ince 2019).

However, the presence of heavy metals in the mortar can harm the strength of the concrete. They delay the hydration of cement and form a low-permeability layer around the unhydrated particles, consequently increasing the hardening time of the concrete. Meanwhile, although heavy metals affect the strength of concrete, there is no standard for their use in concrete. Therefore, to determine the effect of metal mine tailings on the strength of concrete, uniaxial compressive strength (UCS) tests should be performed at various substitution ratios for each tailing, and the results should be compared with the results of the reference sample that contains no tailing.

The chemical content of the material used as aggregate in mortar is also of great importance in terms of mortar strength. For instance, materials with a total of at least 70% SiO_2, Al_2O_3, and Fe_2O_3 content exhibit pozzolanic properties and increase the mortar strength in the long term (Yunhong et al. 2016; Adiguzel et al. 2022). The ability of active silica in the pozzolanic material to react with $Ca(OH)_2$ and water is defined as "pozzolanic activity". With this reaction, strength increases due to the formation of new CSH gels thanks to the pozzolanic material. Meanwhile, CaO in the aggregate dissolves in water and forms a saturated $Ca(OH)_2$ solution that acts as a lubricant/coagulant between the aggregate particles, which changes the plasticity of the mortar. Furthermore, the absorption capacity of lime-forming crystals increases water retention in the mortar (Yunhong et al. 2016; Adiguzel et al. 2022). Therefore, in this study, the basic oxide content of the tailings was revealed via X-ray fluorescence (XRF) analysis; the results are given in Table 2.

Table 2. Basic oxide content of the tailings

Tabela 2. Zawartość tlenków zasadowych w odpadach poflotacyjnych

Basic Oxides	AuT	LZT	CuT	FeT
SiO_2 (%)	42.3	20.1	26.5	49.6
Al_2O_3 (%)	11.2	4.97	7.76	10.8
Fe_2O_3 (%)	7.41	14.2	30.6	30.0
CaO (%)	11.2	45.3	4.87	1.17
K_2O (%)	2.76	1.75	0.38	4.24
MgO (%)	1.69	2.57	3.34	1.01
SO_3 (%)	20.1	9.93	19.5	0.14

As seen in Table 2, the total SiO_2, Al_2O_3, and Fe_2O_3 contents of the tailings, which determine their pozzolanic properties, were 60.91%, 39.27%, 64.86%, and 90.40% for AuT, LZT, CuT, and FeT, respectively. According to these values, it is understood that FeT stood out in terms of the pozzolanic effect but was weak in terms of CaO content, which is the main oxide in supporting cement hydration. Even though the oxides that cause pozzolanic effects in AuT and CuT were below 70%, it can be stated that CaO values will contribute to the binding properties. In this case, it is expected that LZT, which contains 45% CaO, will hydrate faster and give earlier strength to the concrete. In addition, according to the cement standard (EN 197-1), the SO_3 content should be below 4% to prevent concrete from cracking. However, since the aggregate was used as a substitution for aggregate and not for cement in this study, it is necessary to perform UCS tests to understand how these effects will affect the mortar strength. In addition, sulfur or sulfate ions in tailings cause sulfate attacks in concrete. In this case, sulfate ions react with calcium hydroxide and calcium aluminate hydrates, resulting in the formation of gypsum and ettringite. Since the gypsum and ettringite components are large in volume compared to the initially reacting substances, they cause expansion, crack formation, deterioration, and disintegration in reinforced concrete structures (Tuylu et al. 2019). The gypsum is produced as a result of the reaction of calcium and sulfate ions ($Ca^{2+} + SO_4^{2-} = CaSO_4$). In this context, it is seen that FeT had the lowest risk compared to other tailings. Although the SO_3 content of LZT was close to 10%, it had the highest CaO content, which could have a buffer effect against sulfate attacks.

Apart from chemical properties, the physical properties of the aggregates also have a significant influence on the strength of concrete (Shilstone 1990). Therefore, the density, specific surface area, and particle size distribution of the tailings were determined using a helium pycnometer (Ultrapycnometer 1000, USA), a multi-point Brunauer, Emmett, and Teller (BET) analyzer (Autosorb-6, Quantachrome, USA), and a laser particle size analyzer (Mastersizer 2000, Malvern, UK), respectively. Furthermore, the coefficient of uniformity (CU) and coefficient of curvature (CC) values were calculated using Equations (1) and (2), and the results are given in Table 3.

$$CU = \frac{D_{60}}{D_{10}} \tag{1}$$

$$CC = \frac{D_{30}^{\,2}}{D_{10} \cdot D_{60}} \tag{2}$$

The density of the aggregate is an important parameter for its usability in concrete, and it is generally between 2.2 and 2.7 g/cm^3. Aggregates with a density above 3.0 g/cm^3 are considered heavy aggregates, and there is a direct proportion between the mortar strength and aggregate density (EN 12620; Shilstone 1990). In this context, it has been understood that all tailings were adequate for concrete production in terms of density and even suitable for obtaining high strengths.

As the specific surface area of the aggregate increases, it is necessary to increase the cement and water content in the mortar to obtain higher strength and better workability

Table 3. Physical properties of the tailings

Tabela 3. Właściwości fizyczne odpadów poflotacyjnych

Physical property	AuT	LZT	CuT	FeT
Density (g/cm^3)	2.74	3.16	3.38	2.93
Specific surface area (m²/g)	11.59	1.47	11.46	21.72
D_{10} (µm)	2.2	22.9	3.8	104.7
D_{30} (µm)	6.6	69.1	26.3	275.4
D_{50} (µm)	20	120.2	45.7	478.6
D_{60} (µm)	39.8	138	52.5	630.9
D_{80} (µm)	120	182	80	2000
Coefficient of uniformity (CU)	18.09	6.03	13.82	6.03
Coefficient of curvature (CC)	0.5	1.51	3.47	1.15

and consistency, respectively. However, the aggregate cost increases with the concrete amount, while the mortar strength decreases with the amount of water (Carmignano et al. 2021). Therefore, according to the data in Table 3, AuT and CuT-containing mortars may require more water than other tailings. However, fine-grained mine tailings can increase the strength of concrete with a good micro-filling effect and pozzolanic properties depending on their chemical composition (Liang et al. 2017; Sigvardsen et al. 2018). For instance, Moosberg-Bustnes et al. (2004) reported that particle size smaller than 125 µm contributes to the filling effect, while Yunhong et al. (2020) stated that activated siliceous iron tailing has both pozzolanic activity and filling effect. These effects make the internal structure of the concrete more compact and void-free, which can reduce the permeability of the concrete. Consequently, it is considered from Table 3 that LZT is the most convenient tailing for concrete production in terms of its low specific surface area.

It is desired that the aggregate particles in cement mortar exhibit a uniform distribution for an ideal strength. This is possible if the aggregate has a convenient particle size distribution. The appropriateness of the particle size distribution of the aggregate can be determined by calculating the CU and CC values following the standard of geotechnical investigation and testing – identification and classification of soil (EN ISO 14688-2). According to this standard, LZT, CuT, and FeT samples are classified as “medium-graded”, and AuT is classified as “gap-graded” according to their CU values (EN ISO 14688-2).

In addition, the median of the particle size distribution curve (D50) has an important role in expressing the maximum size of half of the total material, along with CU and CC values. The D50 value can also be used to indicate particle size grading. It is expected that there will be a decrease in the void ratio and permeability coefficient values of tailings with “well-graded” particle size distribution.

As a result of the particle size distribution analysis, the size classifications of the tailings were revealed according to the Unified Soil Classification System. Furthermore, methylene blue tests were performed according to the standard of EN 933-9 to determine the clay content of the tailings more accurately. The results are given in Table 4.

Table 4. The size classifications of the tailings according to the unified soil classification system, along with methylene blue values (MBVs)

Tabela 4. Klasyfikacja wielkościowa odpadów poflotacyjnych według ujednoliconego systemu klasyfikacji gleb wraz z wartościami błękitu metylenowego (MBV)

	AuT	LZT	CuT	FeT
Coarse sand size (2000–200 μm) (%)	0	18.5	0	80
Fine sand size (200–75 μm) (%)	30	51	28	7.5
Silt size (75–2 μm) (%)	62	29	67	8
Clay size (<2 μm) (%)	8	1.5	5	4.5
Methylene blue value (MBV) (g/kg)	19.5	1.0	1.75	12.0

Table 4 shows that the tailing with the highest silt content was CuT, while LZT and FeT were rich in fine sand and coarse sand sizes, respectively. Moreover, it is understood from the MBVs given in Table 4 that the lowest MBV of 1.0 g/kg was measured for LZT, which was a tailing that contains limestone as gangue material and contains 45% CaO. Although the clay-sized material content of FeT is close to that of CuT, it is seen that FeT had a considerably higher clay content after AuT, with an MBV of 12 g/kg. This can weaken the bonds between cement and aggregate particles, causing the strength of the concrete to decrease. In this context, the results of Beixing et al. (2011) showed that the performance of concrete containing manufactured sand did not decrease significantly below an MBV of 1.4 g/kg, which can be considered a critical MBV. In another study, Deng et al. (2021) reported that the 3-day compressive strength of concrete was maximized when MBV was around 0.75 g/kg, and the 7-day compressive strength was maximized when MBV was around 1.5 g/kg (Deng et al. 2021). However, MBVs may vary depending on rock types and size ranges. For example, MBVs of limestone aggregates were between 0.3 g/kg and 1.3 g/kg in the presence of 0-2 mm fraction and between 1.7 g/kg and 3.3 g/kg in the presence of 0–0.125 mm fraction. For non-limestone aggregates, MBV values were between 0.3 g/kg and 11.3 g/kg in the presence of 0–2 mm fraction and between 1.7 g/kg and 16.7 g/kg in the presence of 0–0.125 mm fraction (Nikolaides et al. 2007).

The overall results of the physical and chemical characterization indicated that the tailings contain various heavy metals, and this heavy metal content increased their density. Since the high aggregate density has a positive effect on mortar strength, the use of these tailings in concrete production stands out as a good alternative, especially for tailings with lower clay content.

2. Methods

2.1. Preparation of mortars

In this study, mortars without and with tailings to be used in UCS tests were prepared following the standard of "Methods of testing cement – Part 1: Determination of strength" (EN 196-1). Therefore, the mixing type and order were kept constant, as well as the cement type and amount. The amount of water and total mortar were also fixed. Accordingly, the mortars without tailings were prepared using 225 g of water, 450 g of cement, and 1,350 g of standard aggregates and were used as reference samples in the UCS tests. The tailings were used as an aggregate substitution in mortar at various ratios (5–100%) according to the tailing type (EN 450-1; EN 196-1). The prepared mortars were poured into the standard steel molds with the dimensions of 160 mm × 40 mm × 40 mm (length × width × height). Then, the mortar samples were transferred to a water-filled tank at 20 ± 2°C and kept for a total curing time of 28–150 days. The tailing substitution ratios in total aggregate content are given in Table 5 for each tailing along with the curing time used in the experiments (Rajabli 2023).

Table 5. The tailing substitution ratios and curing times of tailing substituted mortar samples

Tabela 5. Współczynniki podstawienia odpadów poflotacyjnych i czasy utwardzania próbek zaprawy z substytucją odpadów poflotacyjnych

	AuT	LZT	CuT	FeT
Tailing substitution ratio in total aggregate content (%)	5	5	5	
	10	10	10	10
	20	20	20	20
		30	30	30
		40	40	40
		50		
		75		
		100		
Curing time (days)	28	28	28	28
	56	56	56	56
	90	90	90	90
	150		150	150

2.2. UCT tests

To determine the effect of the tailing type and substitution ratio on mortar strength and thus the upper limit ratios of the tailing substitution for each tailing type, the samples taken from the tank at the end of the curing period were subjected to UCS tests (Vector, Türkiye) (EN 196-1). In the UCS tests, the force (load) was applied to the placed plates (platens) on the 40 mm × 40 mm (length × width) surfaces of the samples located in the device. The applied force was increased at a constant rate of 2400 ± 200 N/s throughout the test until the concrete sample fractured. The compressive strength (σ_c) of the concrete sample was calculated using the maximum load at fracture as in Equation 3. For each mortar, 5 samples were tested. In order to increase statistical accuracy, the highest and lowest values were not taken into account, and the average value was calculated for the final UCS value.

$$\sigma_c = \frac{F_c}{1600} \tag{3}$$

σ_c – compressive strength (MPa),
F_c – maximum load at fracture (N),
1600 – the area of the platens (40 mm × 40 mm) (mm^2).

The UCS tests were carried out starting from the shortest curing time and the lowest substitution ratio. When the strength value reached a plateau or decreased considerably, it was not deemed necessary to continue the experiments with a higher curing time or substitution ratio.

2.3. pH measurements

Mine tailings can lead to AMD when exposed to atmospheric oxygen and water. As seen in Table 1, the heavy metal content of all tailing samples was above the WHO's upper limit values. In particular, AuT and LZT tailings stand out because of their highest total heavy metal content. Although CuT is close to LZT in terms of the heavy metal content, the reaction of LZT, which has an average SO_3 value, in acidic and basic media was investigated within the scope of this study. Therefore, the effect of SO_3 content as well as heavy metals was revealed. In addition, the SO_3 content of AuT and LZT was ~20% and ~10%, respectively (Table 2). According to these analysis results, AuT and LZT were considered the most hazardous tailings in terms of AMD risk. Therefore, their pH was measured as a function of time and compared with that of AuT and LZT tailings exposed to atmospheric conditions in an open container in the laboratory at 50% moisture and a room temperature of 22±1°C for 30 days, in addition to the mortar samples at maximum tailing substitution ratios of 20% AuT and 100% LZT. Moreover, the pH change of the cement was measured as a function of time.

For this purpose, the samples were dried in an oven (UF 110, Memmert, Germany) at 105°C for 24 h and then ground to below 2 mm using a ring mill (Dinc, Türkiye). Then, their suspensions were prepared at a 20% solid/liquid ratio using pure water (total dissolved solids, TDS = 0–3 ppm) (Libohova et al. 2014). The suspensions were stirred in a magnetic stirrer (YellowLine, IKA, Germany) at a speed of ~500 rpm, and the pH values during the mixing process were measured as a function of time (0–60 min) (Orion Star A215, Thermo Scientific, USA).

3. Results and discussion

3.1. UCS tests

The UCS values of the mortar samples at various tailing substitution ratios are given in Figures 1–4 for AuT, LZT, CuT, and FeT samples, respectively. The graphs also show the strength of the reference concrete made with type I cement (CEM-I) without tailing.

As is known, the concrete produced using CEM-I cement reaches its final strength after 28 days, and increasing the curing time does not create a significant change in the strength of the concrete. As seen in Figure 1, the UCS of this concrete without tailings was measured as 32 MPa in the 28-day curing time, and this value was used as a reference in comparing the UCSs of all mortar samples with tailings.

As seen in Figure 1, the UCS of the concrete sample containing 5% AuT at 28 days is 28 MPa and relatively close to the strength of the reference concrete. The strength increased with the curing time and became equal to the strength of the reference concrete at 60 days.

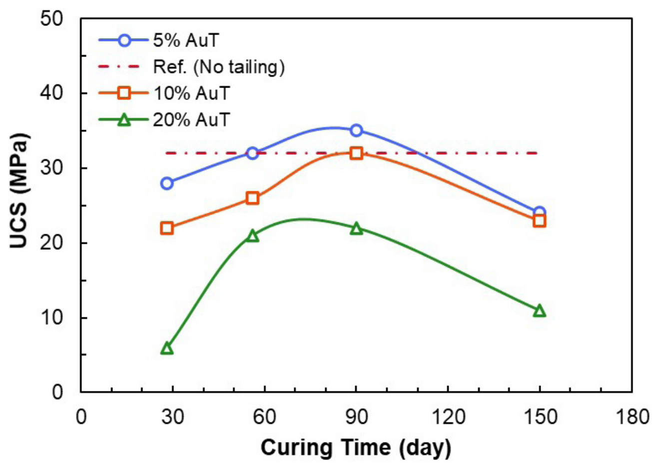

Fig. 1. UCS test results of mortar samples with various AuT substitution ratios

Rys. 1. Wyniki badań UCS próbek zaprawy z różnymi współczynnikami podstawienia AuT

In a 90-day curing time, it reached 35 MPa and exceeded the strength of the reference concrete. The main reason for this increase was the fine particle size of AuT (D_{50} = 20 µm). Fine materials reduced the porosity of the matrix by acting as fillers and creating a denser microstructure. When the AuT ratio increased to 10%, the reference strength value of 32 MPa could be reached in 90 days. It is seen in Figure 1 that AuT provided a pozzolanic effect for up to 90 days. Therefore, when the pozzolanic reaction was combined with the fine particle size of AuT, the compressive strength of the concrete increased. The pozzolanic activity is governed by the chemical composition of the tailings, and the total percentage of SiO_2, Al_2O_3, and Fe_2O_3 should be sufficiently high (min. 70%) (Okonkwo and Arinze 2018). As seen in Table 2, this value is ~61% for AuT.

Due to the high SO_3 content of AuT (~20%), the curing period was extended to 150 days to observe the long-term behavior of the tailing on mortar strength. As seen in Figure 1, at the 150-day curing time, the UCS of the concrete decreased to 24 MPa for 5% AuT and 23 MPa for 10% AuT. The reason for these decreases in strength was the formation of structural cracks along with volumetric expansion due to secondary gypsum and ettringite formations in the concrete due to the high SO_3 in AuT. In addition, since the MBV of AuT was high at 19.5 g/kg, the clay presence can be considered another reason for the decrease in strength.

It is also seen in Figure 1 that mortar strength decreased significantly with the increasing AuT ratio as a general trend. Therefore, the AuT substitution ratio was not increased above 20% in the experimental studies. These results indicated that AuT can be used as an aggregate substitution up to 10%. In accordance with the results of this study, Kunt et al. (2015) reported the optimum gold mine tailing ratio in the concrete as 10%. They stated that a further increase in the tailing ratio affects the mortar strength negatively. On the other hand, according to the results of Ince (2019), the substitution ratio of gold tailings can be increased to 30% thanks to the development of additional calcium silicate-hydrate gels in the presence of tailings.

Concrete is divided into various classes according to its durability and areas of use. In class naming, the number next to the letter C, which symbolizes cement, expresses the minimum compressive strength of 28-day cured concrete in MPa (EN 206-1). High-strength concrete classes such as C50/60 are used in conditions where higher strengths are required, such as large-scale structures.

C30/37-class concretes are preferred in underwater structures due to their high water resistance and dense structures. C20/25-class concrete is the most commonly utilized concrete due to its sufficient strength for general structural elements. C16/20-class concretes with lower strength are generally preferred in non-structural elements to provide a cost advantage. C8/10-class concretes are preferred in ground stabilization and leveling applications.

In this context, considering the fluctuations in the strength of the AuT-substituted mortar samples as a function of curing time, it is seen that AuT tailing can be used in concrete products as non-structural filling materials or those that do not require very high strength, as aggregates in low-strength concrete compositions (C16/20), and as landscape elements such as paving stones, sidewalks, and curb stones in road construction (C8/10).

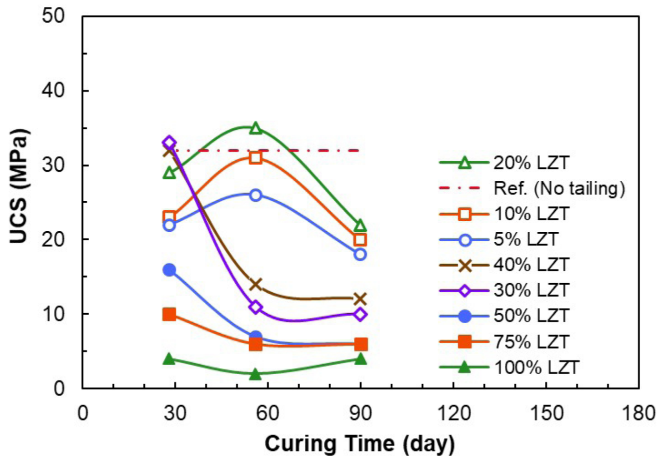

Fig. 2. UCS test results of mortar samples with various LZT substitution ratios

Rys. 2. Wyniki badań UCS próbek zaprawy z różnymi współczynnikami podstawienia LZT

As seen in Figure 2, the UCS of the LZT-substituted mortar samples increased with a substitution ratio up to 30–40% LZT in 28 days of curing time. The highest UCSs in this curing time were obtained at 30% and 40% LZT ratios with 33 and 32 MPa, respectively, which were close to the reference concrete. Increasing the LZT substitution ratio above 40% has a significant negative effect on mortar strength.

In a study conducted by Liang et al. (2017), 40% lead-zinc mine tailing was used as a substitution for aggregate, and it was determined that the UCS value of the concrete with tailing was higher than the UCS value of the reference concrete due to the filling effect (Liang et al. 2017). In another study that used lead-zinc mine tailing as an aggregate substitute, Janković et al. (2017) reported that the UCS of concrete with 5% and 10% substitution was 10% higher than the UCS of the reference concrete at the 28-day curing time. In their study, Janković et al. used sulfate-resistant type III cement (CEM-III) and mineral additive (A mineral additive Type I – limestone). However, in this study, only samples with 30% and 40% LZT substitution were able to exceed the reference value in 28 days.

In the study of Bagger et al. (2021), lead-zinc mine tailing was used for the substitution of cement. In their study, the UCS of the 25% substituted concrete was below that of the reference concrete (Bagger et al. 2021). In the previous studies, which were conducted with lead-zinc tailings, the long-term performance of the concrete was not investigated, and the experiments were stopped after a 28-day curing time. This hinders the detection of conditions such as sulfate attack and clay presence, which may cause strength loss in the long term. To determine whether the tailing contributes to the compressive strength through pozzolanic activity, the curing time should be extended, and strength tests should be applied to these samples (Okonkwo and Arinze 2018).

For this reason, in this study, the strength values at 56 and 90 curing times were also investigated to determine the pozzolanic effects of LZT. When the curing time was extended to 56 days, the UCS of concretes under 20% LZT increased. In this curing time, the UCS approached the reference value with 31 MPa in the presence of 10% LZT and exceeded the reference value with 35 MPa at 20% LZT. A considerable decrease in mortar strength was observed at 30% and 40% LZT. Compared to the UCS values at 28 days, the UCS of concretes with 30% and 40% LZT decreased by almost 60% in 56 days.

When the curing time was extended to 90 days, the UCS of mortar samples under 20% LZT decreased considerably. No significant change was observed in the strength of concretes at higher substitution ratios. It was observed that the increases until the 56th day, which were thought to be a filler effect due to the very fine structure of the material, decreased by ~40% on the 90th curing day. It is considered that the pozzolanic effect did not occur because the total content of SiO_2, Al_2O_3, and Fe_2O_3 in LZT was ~40%. It is also considered that the filling effect caused by the fine size of the material caused an increase in the early UCS values together with ~45% CaO in the tailing, but caused a decrease in the UCS in the long term due to the sulfate attacks, especially originating from the sulfur of the pyrite mineral in the tailing. In this context, the curing time of the LZT samples was not extended further due to the strength decreases measured in 90 days.

Long-term (90-day) strengths of LZT-substituted concretes indicate that concretes containing 5–20% LZT can be used for non-structural elements in C16/20 class, while concretes containing 30–40% LZT can be used as stabilization and leveling materials in C8/10 class.

In Figure 3, it is seen that 30–31 MPa UCS values were obtained in the presence of 5% CuT. However, the strength of the concrete decreased with the CuT substitution ratio. It is seen that this decrease was dramatic above the 20% substitution rate. Therefore,

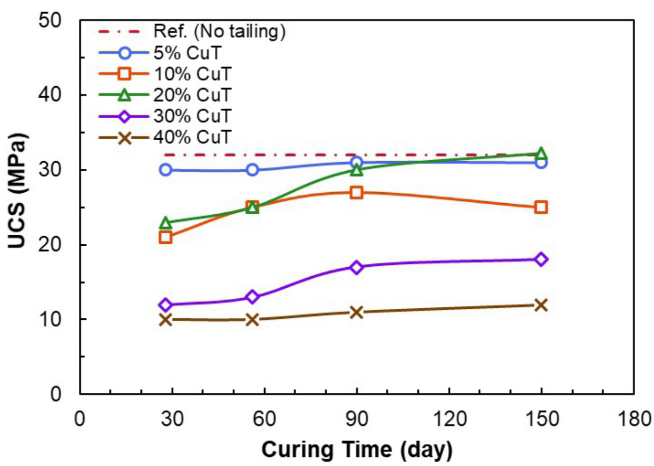

Fig. 3. UCS test results of mortar samples with various CuT substitution ratios

Rys. 3. Wyniki badań UCS próbek zaprawy z różnymi współczynnikami podstawienia CuT it is understood that concretes with a substitution ratio over 20% CuT could not be activated with sufficient cement in 28 days and hence did not gain strength. The curing time increased the mortar strength at all substitution ratios. The strength changes depending on the curing time of the samples with 20% and 30% CuT substitution, showing similar trends. In particular, there was an average increase of 5 MPa in the 56 and 90-day strength values, and an average increase of 1.5 MPa was measured in the following 60-day period. On the contrary, in the samples with 5%, 10%, and 40% CuT substitution, the pozzolanic effect did not occur at a sufficient level, and the expected increase in their strengths did not occur due to the increase in the amount of the tailing. Because the amount of tailing added to the mortar should be at an appropriate rate in order to exhibit pozzolanic behavior together with the cement. This phenomenon can also be seen in Figure 3. In particular, the UCS of concretes containing 20% CuT increased significantly in 56 and 90 days due to the better pozzolanic activity with the binder at these substitution ratios. A similar effect was observed in a study by Pavez et al. (2016) using copper mine tailing as a substitute for aggregate. The authors reported that the UCS of the substituted concrete was above the UCS of the reference concrete at 28-day curing time. In particular, at the end of hydration, obtaining high strengths depends on the cement particles being active, and the activity is achieved by finely grinding the cement (-45 μm) during the production phase. The D_{50} of CuT is also ~45 μm, which can increase the pozzolanic effect in CuT substitution. This effect can also be seen in the 150-day strength values of CuT in Figure 3.

According to the increase in the strength of CuT-substituted concretes with curing time, it is considered that the concretes with 5–20% CuT can be used in the production of general structural elements in the C20/25 class, which is the most widely used.

As seen in Figure 4, the UCSs in 28-day curing time are considerably below the strength of the reference concrete. UCS decreases further with the substitution rate. Meanwhile,

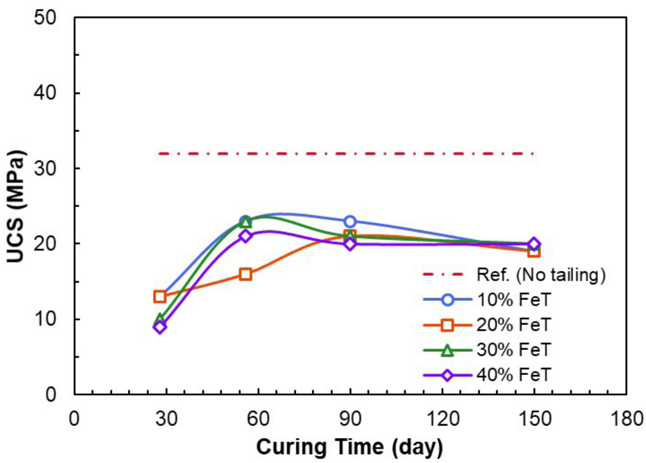

Fig. 4. UCS test results of mortar samples with various FeT substitution ratios

Rys. 4. Wyniki badań UCS próbek zaprawy z różnymi współczynnikami podstawienia FeT an increase was measured in the UCS of the samples after 28 days. This can be explained by the high SiO_2, Al_2O_3, and Fe_2O_3 ratios (~90%) in the FeT content. A similar situation was observed by Zhao et al. (2014), where iron mine tailings were used as a substitute for fine aggregate. The authors reported that the UCS values increased with the curing time.

Despite the increase in UCSs after 28 days, the strength of the tailing-substituted mortar samples remained below that of the reference concrete. Extending the curing time did not cause a significant change in the mortar strength. It is thought that the reason for not obtaining high strengths in the presence of FeT is its high clay content (MBV = 12 g/kg). In addition, it is understood that the filler effect is not formed completely due to the coarse particle size of FeT (D_{50} = 300 μm). In a study by Fontes et al. (2016), iron mine tailings were used as a substitute for aggregate, and it was reported that the concrete containing iron tailings had higher strength than the reference concrete. It is stated that the main reasons for this improvement in mortar strength are the fine particle size distribution and low clay content of the tailings. This opinion is supported by the fact that while the content of the clay-sized material was approximately 2% in the study of Fontes et al. (2016), it was 4.5% for FeT used in this study.

Since the UCS of the FeT-substituted concretes reached a plateau at approximately 20 MPa after 56 days, it is understood that FeT-substituted concretes can be used in C16/20 class, which is generally utilized in the production of non-structural elements.

To compare the effect of the tailing type on mortar strength, UCS results of concretes at tailing ratios where maximum strengths were obtained are given in Figure 5 as a function of curing time, along with the reasons for the change in the UCS.

It is seen in Figure 5 that the concrete with 5% AuT had a higher UCS than the reference concrete (32 MPa), with 35 MPa in 90 days, due to the pozzolanic activity. However, when

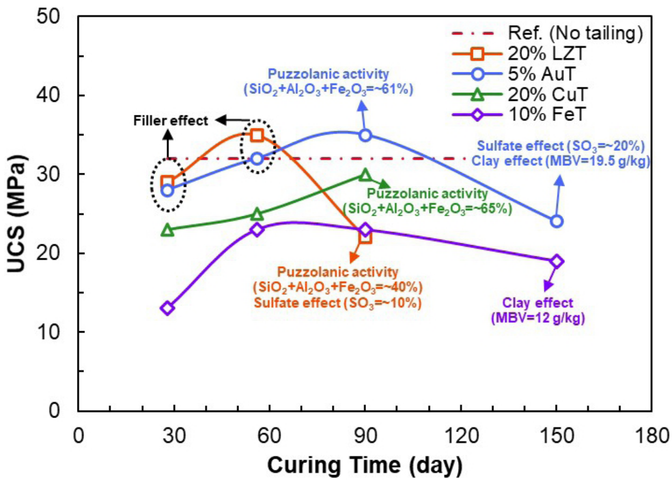

Fig. 5. The highest UCS values of the tailing substituted-concretes, along with the reasons for change in the UCS

Rys. 5. Najwyższe wartości UCS betonów z substytucją odpadów poflotacyjnych wraz z przyczynami zmian UCS the curing period was extended to 150 days, the strength decreased to 24 MPa because of the high sulfur and clay content.

A similar trend is observed in the concrete with 20% LZT. Although it has a higher strength than the reference concrete with 35 MPa in 56 days, the strength values decreased to 22 MPa in 90 days due to the high SO_3 content of LZT.

The UCS of the concrete with 10% FeT reached a maximum of 23 MPa in 56 days, but remained below the reference concrete. Although the SiO_2, Al_2O_3, and Fe_2O_3 contents of FeT are high (90%), the filler effect did not occur in the short term because of its large particle size, and therefore, the strengths remained under the reference concrete. In addition, the high clay content in FeT affected the long-term performance of the concrete negatively.

3.2. pH Measurements

The pH profiles of the AuT and LZT tailings, which are considered environmentally risky, and the samples of these tailings exposed to atmospheric conditions for 30 days are given in Figure 6 as a function of time. Figure 6 also includes the pH profile of the mortar samples in which these tailings were used at maximum ratios (20% AuT and 100% LZT), in addition to the pH profile of cement only.

The measurements were carried out for 60 minutes, and since it was determined that the pH values remained constant for a long time before 60 minutes during the measurements, the experiments were terminated within this period, as also carried out in accordance with the literature (Ali et al. 2020).

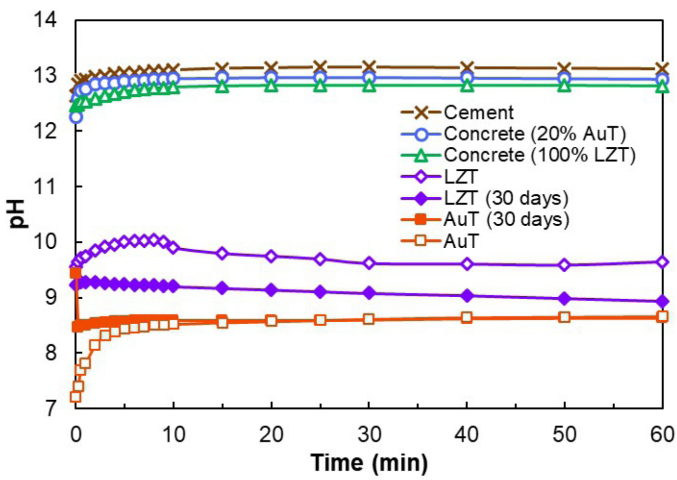

Fig. 6. pH profile of various samples as a function of time

Rys. 6. Profil pH różnych próbek w funkcji czasu

In Figure 6, it is seen that while the pH of AuT in 60 min was 8.66, it was 8.63 for the AuT exposed to atmospheric conditions for 30 days. Meanwhile, the pH of LZT was measured as 9.65 in 60 min, and the pH of LZT exposed to atmospheric conditions for 30 days was 8.94. These results indicated that oxidation did not cause a significant change in the pH level of the tailings.

Figure 6 also shows that the pH values of concrete with 20% AuT and 100% LZT in 60 min were 12.94 and 12.81, respectively, which were highly alkaline. These values were very close to the pH of cement in 60 min (13.12). It is understood that cement significantly increased the pH level of both tailings with high SO_3 and heavy metal content. According to these results, it can be considered that there is almost no AMD risk due to the suppression of the tailings with cement, even in the high sulfur presence.

In accordance with these results, in the study of Kuranchie et al. (2015), where iron mine tailings were used as aggregate substitution material, it was reported that the pH levels of concretes of various ages were between 11.8 and 12.5. These values are quite alkaline conditions and therefore will minimize the potential for acid attack and corrosion, contributing to the concrete's long-term durability. In addition, heavy metals in the tailings will be stored in a stable material such as concrete. Furthermore, Ince (2019) reported that the concentration of heavy metals in the drainage water decreased with the increase in the gold tailings in concrete.

Conclusions

As a result of mining and mineral processing activities, a large amount of mine tailings is produced. These tailings are stored under certain conditions due to technical and environmental reasons, which increases the overall costs of mining activities. Moreover, heavy metal-containing tailings pose an acid mine drainage (AMD) risk when they contact atmospheric water and oxygen.

In light of this information, the use of four different metal mine tailings, namely gold, lead-zinc, copper, and iron, as an aggregate substitution material in mortar was investigated in this study. Therefore, it was aimed to utilize these tailings economically, reduce aggregate consumption, tailing storage costs, and AMD risk, hence increasing the sustainability of the mining activities.

In this context, the effect of the type and ratio of tailing substitution on the mechanical properties (strength) of concrete was investigated via uniaxial compression strength (UCS) tests. Therefore, the maximum ratio at which each type of tailing can be used as an aggregate substitution in concrete was determined. In the study, the environmental impact of the original tailings and tailing-substituted concretes was also investigated with pH measurements as a function of time, thus the risk of AMD formation was revealed.

The results of this study can be summarized as follows:

The UCS of the concrete containing 5% AuT increased with the curing time up to 90 days and reached 35 MPa. The main reason for this increase is the filler effect due to the fine particle size of AuT and the pozzolanic effect through the high SiO_2, Al_2O_3, and Fe_2O_3 content. The strength decreased over 90 days because of the presence of clay and sulfate attacks caused by the high SO_3 content of AuT.

In the presence of LZT, the UCS of the mortar samples increased significantly with the substitution ratio, because of the filler effect, and reached 33 MPa in the presence of 30% LZT in 28 days. However, the strength values decreased with increased LZT ratio and curing time because of the sulfate attacks and low SiO_2, Al_2O_3, and Fe_2O_3 content, which is not enough for the pozzolanic activity. The usability of AuT and LZT tailings in concrete production can be increased by reducing their clay and SO_3 contents.

In the presence of 5% CuT, the UCS of the concrete was quite close to that of the reference concrete with 31 MPa. The substitution ratio had a negative effect on the strength of the concrete, but the strength values can be increased with curing time, depending on the substitution ratio. For instance, 30 MPa strength was obtained in 90 days in the presence of 20% CuT.

In the presence of FeT, the UCS of the mortar samples remained below the reference concrete. Although there was an increase in strength after 28 days due to the pozzolanic effect, the final strength values were around 20 MPa. If the particle size and the clay content of FeT are reduced, higher mortar strengths can be obtained.

As a result of mechanical investigations carried out in this study, it can be stated that metal mine tailings can be used as a partial aggregate substitution in concrete production between the ratios of 5% and 20%, depending on the tailing type. The UCS test results also showed that, depending on the substitution ratio, AuT, LZT, and FeT substituted concretes can be used in the construction of non-structural elements in the C16/20 class, while the strength of CuT substituted concrete can reach up to the C20/25 class.

In the environmental approach, the results of the pH measurements carried out in this study indicated that the pH values of AuT and LZT, which are the tailings with the highest potential for AMD formation, are approximately between 8.5 and 9.5 and do not change significantly due to atmospheric oxidation. Therefore, the process after the direct contact of atmospheric oxygen with the tailing, which is necessary for the formation of AMD, as well as sulfur and moisture, was investigated. When these tailings are used as aggregate substitution material in concrete production, the pH of the produced concrete approaches 13 due to the presence of cement. At these highly alkaline pH values of the concrete, there is almost no risk of the tailings forming AMD.

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The Authors have no conflicts of interest to declare.

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A MECHANICAL AND ENVIRONMENTAL APPROACH ON THE PARTIAL SUBSTITUTION FOR AGGREGATE IN MORTAR USING VARIOUS METAL MINE TAILINGS

Keywords

sustainable mining, mortar, uniaxial compression strength (UCS), acid mine drainage (AMD), methylene blue value (MBV)

Abstract

The problem of managing the high amounts of mine tailings has become important with the increase in production in mining activities. The tailings must be stored or disposed of in a controlled manner to eliminate or minimize their negative effects such as air, water, and soil pollution. In particular, the tailings of metal mines have acid mine drainage (AMD) risk and must be stored under safe conditions. The use of these tailings in an appropriate industry, such as concrete production, will increase the sustainability of mining activities. This study investigates the effect of gold (AuT), lead-zinc (LZT), copper (CuT), and iron (FeT) metal mine tailings as a partial substitution base material for aggregate in concrete in terms of mechanical and environmental aspects using uniaxial compression strength (UCS) tests and pH measurements, respectively. The results indicated that the strength of the reference concrete without tailing (32 MPa) could be reached or even exceeded by tailing substituted mortar samples with 5% AuT, 5% CuT, and 20% LZT. The strength of several mortar samples increased with the curing time due to the filler effect and pozzolanic activity, while it decreased due to the clay effect and sulfate attacks depending on the tailing type and the substitution ratio. The pH value of the tailings, which was between 8.5 and 9.5, was not affected significantly by atmospheric oxidation. When these tailings were used in concrete production, the suspension pH approached 13 due to the alkaline properties of the cement, almost eliminating the AMD risk.

MECHANICZNE I ŚRODOWISKOWE PODEJŚCIE DO CZĘŚCIOWEGO ZASTĄPIENIA KRUSZYWA W ZAPRAWIE Z WYKORZYSTANIEM RÓŻNYCH ODPADÓW POFLOTACYJNYCH Z KOPALNI METALI

S ł o w a k l u c z o w e

zrównoważone górnictwo, zaprawa, wytrzymałość na ściskanie jednoosiowe (UCS), kwaśny drenaż kopalniany (AMD), wartość błękitu metylenowego (MBV)

S t r e s z c z e n i e

Problem zarządzania dużymi ilościami odpadów poflotacyjnych stał się istotny wraz ze wzrostem produkcji w górnictwie. Odpady poflotacyjne muszą być składowane lub utylizowane w sposób kontrolowany, aby wyeliminować lub zminimalizować ich negatywne skutki, takie jak zanieczyszczenie powietrza, wody i gleby. W szczególności odpady poflotacyjne z kopalni metali stwarzają ryzyko kwaśnego odwodnienia kopalni (AMD) i muszą być składowane w bezpiecznych warunkach. Wykorzystanie tych odpadów w odpowiednim przemyśle, takim jak produkcja betonu, poprawi zrównoważony rozwój działalności górniczej. W niniejszym badaniu zbadano wpływ odpadów kopalnianych złota (AuT), ołowiu i cynku (LZT), miedzi (CuT) i żelaza (FeT) jako częściowego substytutu kruszywa w betonie pod względem aspektów mechanicznych i środowiskowych, wykorzystując odpowiednio testy wytrzymałości na ściskanie jednoosiowe (UCS) i pomiary pH. Wyniki wskazały, że wytrzymałość betonu referencyjnego bez odpadów (32 MPa) można osiągnąć lub nawet przekroczyć, stosując próbki zaprawy z 5-procentowym dodatkiem AuT, 5-procentowym dodatkiem CuT i 20-procentowym dodatkiem LZT. Wytrzymałość kilku próbek zaprawy wzrosła wraz z czasem utwardzania ze względu na efekt wypełniacza i aktywność pucolanową, podczas gdy spadła ze względu na efekt gliny i ataki siarczanów, w zależności od rodzaju odpadów i współczynnika podstawienia. Wartość pH odpadów, która wynosiła od 8,5 do 9,5, nie została znacząco zmieniona przez utlenianie atmosferyczne. Po wykorzystaniu tych odpadów w produkcji betonu pH zawiesiny zbliżyło się do 13 ze względu na zasadowe właściwości cementu, co praktycznie wyeliminowało ryzyko AMD.