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TECHNICAL ARTICLE

The Reuse of Gold Tailings as Fill Material for Depleted Mines

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Abstract

The accumulation of gold mine tailings poses an environmental challenge. The feasibility of reusing this waste to fill underground mines was investigated. The tailings were mixed with Portland cement and water using a one-step method. The fluidity, bleeding rate, setting time, mechanical behavior, water quality analysis, and toxicity characteristics of the leachates with various tailings/cement ratios and slurry concentrations were characterized and assessed. The results show that secondary hydration reactions occurred between the tailings and the cement. The rheological properties, setting/hardening properties, and bleeding rate became less favorable with increases in the tailings/cement ratio and slurry concentrations. The uniaxial compressive strength of the filling materials prepared with a tailings/cement ratio of 4:1 and a slurry concentration of 68% was 0.99 MPa, which met the filling purposes. The water quality index of the leaching solution was acceptable, with the concentrations of copper (Cu^{2+}), zinc (Zn^{2+}), cadmium (Cd^{2+}), lead (Pb^{2+}), total chromium (Cr^{3+} and Cr^{6+}), hexavalent chromium (Cr^{6+}), beryllium (Be^{2+}), barium (Ba^{2+}), silver (Ag^{+}), selenium (Se^{2-}), nickel (Ni^{2+}), mercury (Hg^{2+}), arsenic (As^{2+}), fluoride (F^{-}), and cyanide all much less than the standard limits. This indicates that gold mine tailings could be used as fill material to reduce waste accumulation without causing any hazards to the environment or human beings.

Keywords Gold mine tailings · Compressive strength · Fluidity · Leaching solution

Introduction

Gold mine tailings are an industrial waste left after the beneficiation or recovery of valuable elements from gold ore. Approximately 38.3 tons of tailings are generated for every ton of refined gold produced (Feng et al. 2018). Sichuan is a major province for mineral resources and the amount of industrial solid waste, including tailings, has been gradually increasing. In 2019 and 2020, more than 90 million cubic meters of tailings were accumulated in the province, with a comprehensive utilization rate of only 27.7%. Among them, the utilization rate of gold mine tailings was approximately 26% (Wang et al. 2022a). The arbitrary disposal of tailings increases environmental risks and wastes resources. Therefore, it is essential to find practical ways to address this problem.

Previous research on the comprehensive utilization of tailings includes the recovery of residual gold and associated metal elements such as Ag, Cu, Pb, Zn, and W (Li et al. 2017; Mostafa and Abdullah 2019; Peelman et al. 2018; Wang et al. 2019) and non-metallic minerals such as quartz, feldspar, and sericite (Ahmed et al. 2021; Wei et al. 2014). However, in recent years, the gold grade of mines and the value of tailings has gradually decreased. In addition, the purification of quartz and recovery of feldspar and sericite would increase the amount of waste (Cairncross and Tadie 2022). Reclamation of most large tailings piles requires a large investment, as it must both restore the land use value and the ecological environment (Compaore et al. 2020; Fang et al. 2021; Wang and Chen 2020; Xia et al. 2020; Zhou et al. 2019).

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Gold tailings have a high SiO_2 and Al_2O_3 content, and contain CaO, MgO, Na_2O, K_2O, and other components that are conducive to cemented filling and use as underground backfill material (Ahmed et al. 2021; Behera et al. 2021; Cairncross and Tadie 2022; Chen et al. 2022, 2023; Liu et al. 2023; Mostafa and Abdullah 2019; Peelman et al. 2018; Saedi et al. 2023; Wang et al. 2019; Wei et al. 2014; Zhang and Zhang 2021). This use of solid waste to fill mine voids can solve the problems of deformation and subsidence caused by underground mining and has become the most widely used method in the mining fields in recent years (Ahmed et al. 2021; Deng et al. 2017; Mostafa and Abdullah 2019; Wang et al. 2019).

Wang et al. (2019) found that the concentration of the slurry and the tailings/cement ratio of the slurry were respectively the most important and second-most important factors in dictating the compressive strength of the filling materials. The concentration of the slurry was given by:

$$Concentration = \left[m_{Tailings} + m_{Cement}\right] / \left[m_{Tailings} + m_{Cement} + m_{Water}\right] \quad (1)$$

where $m_{Tailings}$, m_{Cement}, and m_{Water} represent the mass of tailings, cement, and water, respectively. Furthermore, they used tailings from the no. 4 mine in Shandong Province, with a ratio of tailings to fly ash of 1:6 and a slurry concentration of 76% (Wang et al. 2022a). The compressive strength of the filling materials after 3 days, 7 days, and 28 days were extrapolated to be 0.99, 2.48, and 4.39 MPa, respectively, which met the compressive strength requirements of the backfill materials for mined-out areas. The addition of different types, strengths, and dosages of cement markedly affected the compressive strength of the filling material. The compressive strength of the filling materials decreased as the ratio of cement to tailings decreased. To date, many papers have shown that the compressive strength of the filling materials significantly improves over time and can meet the compressive strength requirements of backfill materials (Behera et al. 2021; Chen et al.2022, 2023; Deng et al. 2017; Liu et al. 2023; Li et al. 2019; Saedi et al. 2023; Tebogo and Thandiwe 2021; Wang et al. 2022b; Xu et al. 2013; Yin et al. 2012; Zhang and Zhang 2021). However, good working performance of the filling slurries is essential for the backfill process and there is limited research on the working performance of these slurries (Mardani-Aghabaglou et al. 2021).

It is equally important to pay attention to the leaching toxicity of the backfill materials (Fredy et al. 2021). When contaminants are leached by surface water or groundwater after backfilling, hazardous components can be released into the environment (Chen et al. 2021; Hamberg et al. 2017; De Oliveira Ribeiro et al. 2020; Mohammadali et al. 2023; Olías et al. 2021; Sibele et al. 2023). Various methodologies have been used to explore the leaching potential of backfill materials. Metals (including copper (Cu^{2+}), arsenic (As^{2+}), zinc (Zn^{2+}), cadmium (Cd^{2+}), lead (Pb^{2+}), total chromium (Cr^{3+} and Cr^{6+}), silver (Ag^{+}) and nickel (Ni^{2+})) leaching from backfill materials has been tested by Saedi et al. (2023), Sibele et al. (2023), Hamberg et al. (2017), Chen et al. (2021).

However, there is lack of information on the amount of total phosphorus, ammonium nitrogen, chemical oxygen demand(COD) and fluoride the cured slurries would release to water bodies, as these chemicals would cause secondary pollution to the environment. In addition, the workability of the slurries has rarely been reported. Based on previous research, gold tailings were used as the main substance and Portland cement was used as binding media to prepare filling materials for the mined-out areas. The influence of different slurry concentrations and tailings-cement ratios on fluidity, bleeding (water seepage) rate, setting time, mechanical properties, water quality and toxicity of leaching solution of the filling materials were investigated, in order to obtain the optimal amount of tailings in the filling materials.

Materials and Methods

Materials

Gold mine tailings were collected from the Dabaoshan gold mine in Pingwu, Mianyang City, Sichuan Province. The chemical composition, particle size, density, water content, and other physical properties of the tailings are shown in Tables 1, 2, 3 and 4. According to Table 1, the tailings contained very high amounts of SiO_2 content and high amounts of Al_2O_3. The chemical composition was similar to ordinary construction materials. The gold mine tailings samples contained 0.30 g/t Au, based on elemental analysis (Table 2). It can be seen from Table 3 that the overall particle size of the tailings was very small. The retention rate of -0.043 mm particle size in the tailings was 52.19%, and the retention rate of -0.074 mm particle size reached 21.06%. If not properly handled, the tailings can cause hazards to the surrounding environment, such as soil erosion and dust. Chuanxiong Cement P.O42.5R, produced by Sichuan Chuanxiong Building Materials LLC in Deyang City, Sichuan province was used as the primary binder. Tap water was used for preparing the filling materials.

From Table 1, according to formulas 2 and 3

Table 1 Oxide composition of the gold mine tailings (oxide based ore, no sulphide)

Chemical combination	SiO_2	CaO	Fe_2O_3	Al_2O_3	MgO	TiO_2	P_2O_5	Na_2O	K_2O
Wt(%)	60.88	0.777	7.07	20.05	1.15	0.625	0.0905	1.92	4.22

Table 2 The elemental analysis of the gold mine tailings (weight content)

Element	Au*	Zn	Cd	Pb	As	S
Wt(%)	0.30	0.0062	0.00039	0.00146	0.00132	0.0567
Element	Ag*	Se	Ni	Cu	Cr	WO_3
Wt(%)	< 5	0.000006	< 0.00010	0.00042	0.01315	0.0202

*Content's unit is g/t

Table 3 Particle size, mass, grader percentage and cumulative percentage of the gold mine tailings

Particle size (mm)	Mass (g)	Grader percentage (%)	Cumulative percentage (%)
+ 0.25	5.5	2.77	2.77
− 0.25 + 0.1	32.1	16.17	18.94
− 0.1 + 0.074	15.5	7.81	26.75
− 0.074 + 0.043	41.8	21.06	47.81
− 0.043	103.6	52.19	100.00
Total	198.5	100.00	/

$$M_0 = [m(\mathrm{CaO}) + m(\mathrm{MgO})] / [m(\mathrm{SiO_2}) + m(\mathrm{Al_2O_3})] \tag{2}$$

$$A_0 = [m(\mathrm{CaO})] / [m(\mathrm{SiO_2})] \tag{3}$$

where M_0 represents the alkaline coefficient of the tailings, A_0 is the activity coefficient, and $m(CaO)$, $m(MgO)$, $m(SiO_2)$, and $m(Al_2O_3)$ indicate the mass content of CaO, MgO, SiO_2, and Al_2O_3, respectively. The alkaline coefficient of the tailings ($M_0 = 0.024$) was less than 1, and the activity coefficient of the tailings A_0 was 0.013, suggesting that the tailings were acidic and inert (Deng et al. 2017).

Preparation

The filling materials were prepared using a one-step method (see Fig. 1); the raw tailings were mixed directly with cement and water, without any preprocessing, such as separation or purification. It was a simple low-cost process. The mass ratios of the tailings, cement, and water for each specimen are listed in Table 5. The tailings and cement were mixed thoroughly by hand to ensure even distribution. Water was added to the above mixture and stirred by different protocols of rotation speed using an electric mixer (Beijing Zhongjiao Construction Instrument Technology Development Co.LTD, Fangshan District, Beijing, China). The slurries were then poured into standard cylindrical molds with a diameter of

Table 4 Physical properties of the gold mine tailings

Moisture content (wt%)	Density		Porosity (%)	Void ratio	Saturation (%)	Specific weight of soil particle	Quick shear test	
	Dry density (g/cm^3)	Wet density (g/cm^3)					Cohesion	Internal frictional angle
24	1.35	1.20	57.6	1.358	25.30	2.84	24.0	43.6

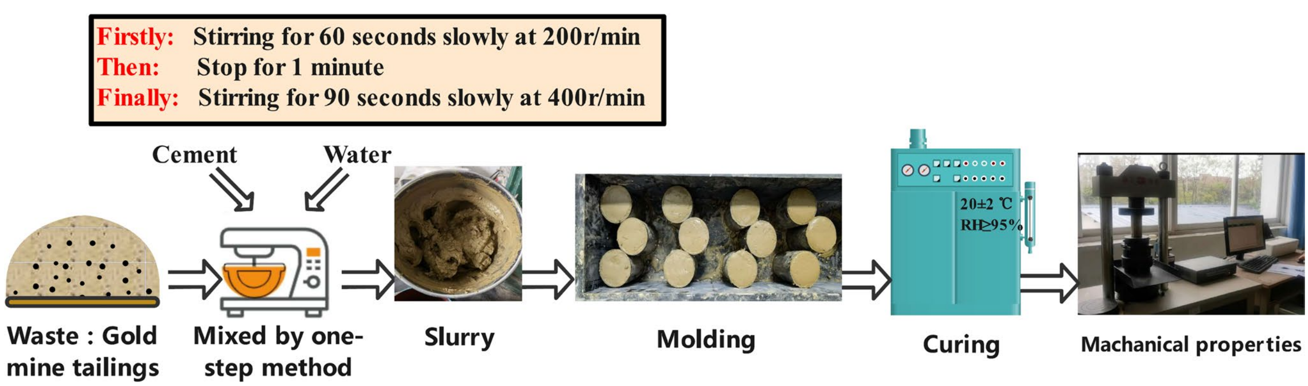


Fig. 1 The diagram showing the preparation of the filling materials

Table 5 Slurry concentration and tailings/cement ratio of all specimens

No	Slurry concentration	Tailings/cement ratio
1	68%	4
2		6
3		8
4		10
5		12
6	72%	4
7		6
8		8
9		10
10		12
11	76%	4
12		6
13		8
14		10
15		12

5 cm and a height of 10 cm. After standing for 3 min, the surface of the specimens was leveled. A total of 45 specimens were prepared and placed in a standard curing chamber with a humidity ≥ 90% and temperature of 20 ± 1 °C until the required age was reached.

Characterizations

The flowability was tested according to GB/T2419-2005 (the method for determining the flowability of cement mortar). The slurry was prepared and divided into two layers in a truncated cone mold produced by Zhongjian Precision Instrument Co., LTD (Cangzhou, Hebei Province, China), with a height of 60 ± 0.5 mm, an upper inner diameter of 70 ± 0.5 mm, a lower inner diameter of 100 ± 0.5 mm, and a lower outer diameter of 120 mm. After the slurry was compacted and placed on a vibration table, the mold sleeve was taken off and the surface of the slurry was smoothed gently. Then the table was programmed to deliver 25 vertical oscillations. The flow diameters were then measured in two perpendicular directions using a caliper.

Setting time was measured according to GB/T1346-2011 (the method for determining the water demand, setting time, and stability of cement standard slurry). The slurry was loaded into a round mold as required. After curing for 30 min, the sinking depth of the test needle was measured every 5 min. The initial setting time was reached when the sinking depth of the test needle from the bottom plate was 4 ± 1 mm. To measure the final setting time, the specimen was turned over and a new test needle was used. The sinking depth of the new needle was measured every 15 min. The final setting time was reached when the sinking depth of the needle was 0.5 mm.

The bleeding rate was measured according to GB/T50080-2016 (the test method for properties of ordinary concrete mixtures). The slurry was loaded and compacted into a cylinder (with a diameter of 108 mm and a height of 109 mm). The surface of the slurry was smoothed and was 10 mm below the mouth of the cylinder. The bleeding of the slurry was collected every 10 min during the first 60 min. After that, the bleeding was collected every 30 min until there was no more bleeding. A 35 ± 5 mm thick cushion block was padded under the cylinder 2 min ahead of bleeding collection to ensure that water was fully excreted. The collected water was placed in a measuring cylinder with a cap and the amount of water was recorded after each bleeding collection. Finally, the accumulated bleeding was calculated.

A uniaxis compression test was conducted on the specimens that had reached the required age using an EHC-1300 microcomputer-controlled electronic universal testing machine (Modern Instrument, Shanghai, China), with a compression rate of 0.5 kN/s. To reduce errors, three specimens were measured for each group and each age, and the average value was taken as the final compressive strength of the filling materials.

The morphology of all specimens was studied by a 7800F field emission scanning electron microscope (FE-SEM, JEOL, Tokyo, Japan). Fractured pieces (collected from samples after the compression test) were used as specimens for the SEM investigation. The specimens were soaked in 99% anhydrous ethanol for 24 h to prevent hydration and then subjected to a thin layer of gold coating before SEM investigation. The gold-coated specimens were loaded into the SEM to observe the surface morphology.

The crystal structures of the specimens were investigated using an Empyrean diffractometer, which is a third-generation x-ray diffractometer (XRD) manufactured by Malvern Panalytical Ltd (Malvern, UK). The rotary half-cone angle 2θ was set from 10 to 90° with 2°/min of scanning velocity and 0.02 of scan step angle.

For water quality analysis of the leachate, distilled water was used as the extractant to simulate the process of components being leached out of the filling materials by surface water or groundwater. About 20 g of the cured tailings fill material was weighed at an age of three days and placed in a transparent container, and five times the amount of water was added to fully saturate the tailings fill material in the container. After seven days, the liquid was taken out for tests. The concentration of total phosphorus, ammonia nitrogen, chemical oxygen demand (COD), suspended solids, fluoride, and pH were determined by ammonium molybdate spectrophotometry, Nessler's reagent spectrophotometry, rapid digestion spectrophotometry, filter paper, ion chromatography, and calibrated digital pH meter, respectively. The spectrophotometer used in those experiments was produced by CANY Precision Instruments Co., Ltd in Shanghai, China. The filter paper was purchased from Newstar Paper Co., Ltd in Hangzhou, China. The 925 CN Ion chromatography used in the work was manufactured by Metrohm in Swiss. The pH was measured using a PHS-25 digital acidity meter (Shanghai INESA Scientific Instrument CO. Ltd, Shanghai, China).

The metals in the leachate were also tested with distilled water; 100 g of distilled water and 20 g of the backfill materials were placed into a 200 ml beaker and then left to stand for 14 days. The leachate was analyzed for the concentrations of copper (Cu^{2+}), zinc (Zn^{2+}), cadmium (Cd^{2+}), lead (Pb^{2+}), total chromium (Cr^{3+} and Cr^{6+}), hexavalent chromium (Cr^{6+}), beryllium (Be^{2+}), barium (Ba^{2+}), silver (Ag^{+}), selenium (Se^{2-}), nickel (Ni^{2+}), mercury (Hg^{2+}), arsenic (As^{2+}), fluoride (F^{-}) and cyanide.

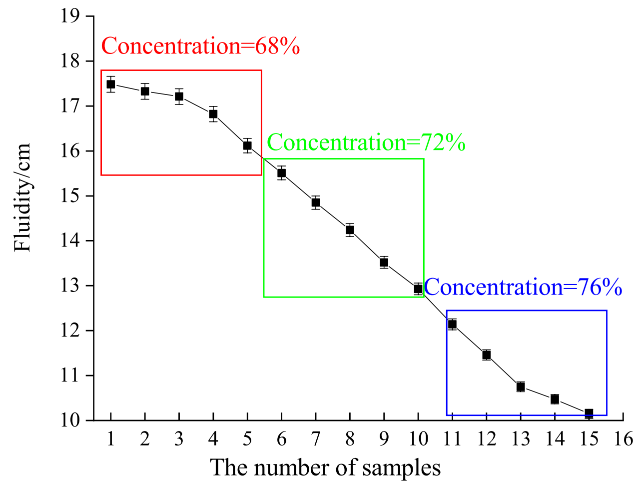


Fig. 2 The effect of concentration and tailings/cement ratio on the fluidity of all the specimens

Results and Discussion

The influence of slurry concentration and tailings/cement ratio on the performance of gold mine tailings fill materials were investigated and assessed. Fifteen sets of experiments were designed with different tailings/cement ratios (4, 6, 8, 10, and 12) and concentrations (68%, 72%, and 76%). The properties of the filling materials were investigated, including flowability, setting time, bleeding rate, mechanical properties, and water quality of leaching solution.

Flowability and Setting Time Tests

The flowability of the tailings fill material is an important factor for the grouting process. Generally, excellent flowability of the slurry leads to a smooth grouting filling process (Deng et al. 2017). As shown in Fig. 2, the flowability of the tailings fill material decreased as the ratio of the tailings mixture to water was increased. A higher tailings/cement ratio also resulted in less flowability. The first is because an increase in the concentration led to a decrease in the amount of water used, while an increase in the tailings/cement ratio reduced the amount of cement paste produced, resulting in less lubrication and a drier slurry (see Fig. 3). In addition, the increased amount of tailings had more surface areas that need to be wetted.

Setting time is also an important parameter in the grouting filling process (Yin et al. 2012). A suitable setting time ensures that there is enough time to complete the construction requirements and that the slurry can harden in time to resist groundwater erosion (Xu et al. 2013). As shown in Table 6, when the concentration was 68% and the tailings/cement ratio was 4:1, the setting time of the material was relatively short, with an initial setting time of 7 h and a final setting time of 12 h.

The setting time of the fill materials increased with an increase of the tailings/cement ratio. When the tailing/cement ratio was between 6 and 12, the cement content decreased, and the setting time of the slurries became longer. Furthermore, both the initial and final setting times decreased as the tailings/cement ratio increased. A similar conclusion was obtained when the concentration of the slurries was 72%.

Bleeding Measurements

The results for the bleeding measurement (water seepage rate) of the tailings filling slurries are shown in Fig. 4. With an increase of the concentration and the tailings/cement ratio, the slurries gradually became drier and the bleeding rate of the filling materials decreased, which agrees with the flowability results. This was because when the concentration was relatively low (i.e. 68%), the content of the solid material (tailings and cement) was low and the amount of water used was high. Since the amount of water used in the hydration of cement was fixed, the excess water separated out. When the tailings/cement ratio was 8, the amount of cement added was greatly reduced, resulting in more water being separated out. A similar conclusion was obtained when the concentration of the slurries was 72%. However, the amount of cement paste formed was the least when the concentration of the slurries was further increased to 76%; therefore, the lubricating effect of the cement paste was not obvious.

Fig. 3 The slurry with: **a** a concentration ≤ 72% and **b** a concentration = 76%

Table 6 The influence of concentration and tailings/cement ratio on the setting time of all the specimens

Number	Concentration	Tailings/cement ratio	Initial setting time (h)	Final setting time (h)
1	68	4	7 ± 0.07	12 ± 0.33
2		6	8 ± 0.13	24 ± 0.5
3		8	5 ± 0.08	20 ± 0.47
4		10	5 ± 0.06	20 ± 0.42
5		12	5 ± 0.11	20 ± 0.5
6	72	4	4 ± 0.06	5 ± 0.33
7		6	4 ± 0.07	5 ± 0.12
8		8	4 ± 0.08	5 ± 0.13
9		10	2 ± 0.05	4 ± 0.08
10		12	2 ± 0.04	4 ± 0.11
11	76	4	N/A, too dry(Fig. 3b)	N/A
12		6	N/A	N/A
13		8	N/A	N/A
14		10	N/A	N/A
15		12	N/A	N/A

Mechanical Properties

The variation of the uniaxial compressive strength of tailings filling materials is shown in Fig. 5. It can be seen from Fig. 5a that under the same concentration, the uniaxial compressive strength of the filling materials gradually decreased with an increase in the tailings/cement ratio, indicating that the development of strength in the system was still dominated by the hydration of cement. When the tailings/cement ratio was fixed at 4:1, with the increase of the concentration, the compressive strength of the filling material gradually decreased as the concentration decreased (Fig. 5b). However, the uniaxial compressive strength of the filling materials shows a turning point when the tailings/cement ratio was ≥ 6:1, the compressive strength of the materials with higher concentration is greater than those of materials with lower concentration. This is because with the increase of the concentration, the amount of water decreased and the slurry became drier, which is in agreement with the flowability results. The cement was fully hydrated, and the alkaline and salt excitations for the secondary hydration reaction of tailings were better, resulting in better mechanical properties. In summary, the optimal uniaxial compressive strength of the filling materials was achieved when the concentration of the materials was 68% and the tailings/cement ratio was 4:1.

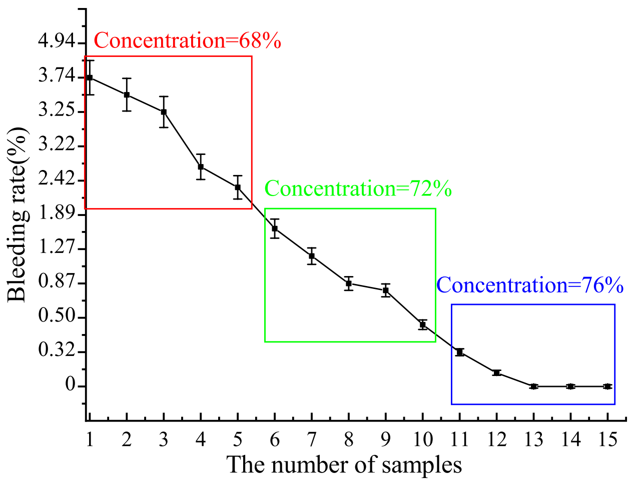


Fig. 4 The bleeding rate of all the specimens

Quality Analysis of the Leaching Solution

Quality analysis of the leaching solution, shown in Table 7, has not been reported in any former literature. The levels of ammonia nitrogen, COD, and fluoride were much less than the level I standard of GB 8978-1996 (comprehensive wastewater discharge standard, in which the limits of ammonia nitrogen, COD, and fluoride are 15 mg/L, 100 mg/L, and 10 mg/L, respectively). In addition, the level of phosphate was less than 1.0 mg/L according to the level II standard. The COD, ammonia nitrogen, and pH values of the leaching solutions also met the class V groundwater quality standards of GB14848-2017 (the limits are: COD > 10 mg/L, ammonia nitrogen > 1.5 mg/L and pH > 9.0). The fluoride ≤ to 2.0 mg/L, which conforms to class IV of GB14848-2017. Nevertheless, the data on suspended solids and phosphate are not specified. Since the chemical content of the groundwater is high, the wastewater cannot be used as drinking water, but can be used for industrial purposes.

Leaching Toxicity Measurement

Potentially, the leaching of hazardous components from gold mine tailings-containing filling materials, prompted by surface water or groundwater movement following backfilling, might lead to the release of these substances into the environment, consequently giving rise to secondary pollution. Hence, safe management and accurate reuse of tailings becomes imperative. Table 8 presents the results of metal leaching of sample 1 and the leaching standard based on GB 5085.3 2007 (identification standards for hazardous wastes, identification for extraction toxicity).

The concentrations of copper, zinc, barium, and fluoride were measured at 0.002, 0.0059, 0.0012, and 0.78 mg/L, respectively, which were much less than the standard limited value of 100 mg/L. Analogously, the amounts of lead, hexavalent chromium, argentum, nickel, arsenic, and cyanide leaching were less than the standard limit (5 mg/L). The cadmium content in the sample was 0.001 mg/L, while the standard limit is 1 mg/L. The measured value of total chromium was only 1/10,000 of the standard value and the selenium content was 1/20,000 of the standard value. The beryllium and mercury concentrations in the sample were 0.00056 and 0.00026 mg/L, respectively. According to GB 5085.3 2007 (the identification standards for hazardous wastes, identification for extraction toxicity), the maximum amounts of these two components are 0.02 and 0.1 mg/L, respectively.

Following a 14-day curing period, the metal concentrations were notably below the standard limits. Meanwhile, as the curing days increased, the advanced hydration process resulted in greater stabilization of the hazardous components. In addition, extending the curing period allowed the calcium silicate hydrate (C–S–H) gel to gradually occupy the pores, leading to increased cohesion and consequently enhanced stabilization of metals (Behera et al. 2021). Therefore, gold mine tailings can be used as backfill materials without causing environmental concerns.

Fig. 5 the uniaxialcompressive strength of the filling materials as a function of: **a** ratio of tailings/cement and **b** concentration

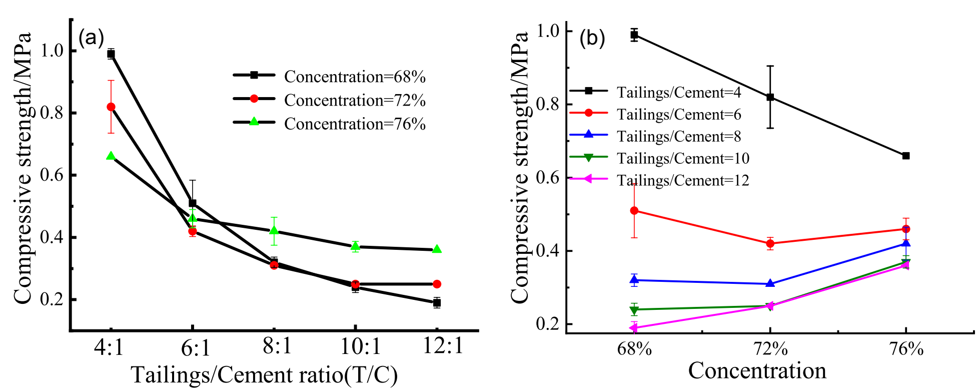


Table 7 The water quality analysis of leaching solution

NO	c	T/C	pH	LV_{pH}	SS (g/L)	LV_{SS}	COD (mg/L)	LV_{COD}	P (mg/L)	LV_P	AN (mg/L)	LV_{AN}	F (mg/L)	LV_F
1	68	4	12.14 ± 0.01	I: 6–9	1.06 ± 0.004	I: 100	10.18 ± 0.001	I: 100	0.86 ± 0.003	I: 0.5	0.57 ± 0.00	I: 15	0.94 ± 0.003	I: 10
2		6	12.17 ± 0.01	V: pH < 5.5	0.91 ± 0.002	N/A	11.76 ± 0.001	V: > 10	0.36 ± 0.001	II: 1.0	1.22 ± 0.004	V: > 1.50	1.00 ± 0.001	V: > 2.0
3		8	12.16 ± 0.01	or pH > 9.0	0.88 ± 0.000		10.97 ± 0.000		0.67 ± 0.001	N/A	1.92 ± 0.002		1.37 ± 0.004	IV: ≤ 2.0
4		10	12.26 ± 0.02		0.76 ± 0.002	±	13.34 ± 0.004		0.30 ± 0.001		0.60 ± 0.001		0.66 ± 0.001	
5		12	12.13 ± 0.01		1.07 ± 0.002		14.93 ± 0.004		0.47 ± 0.005		0.38 ± 0.004		0.70 ± 0.001	
6	72	4	12.25 ± 0.02		0.80 ± 0.004		14.93 ± 0.004		0.54 ± 0.003		0.21 ± 0.003		0.59 ± 0.003	
7		6	12.32 ± 0.02		1.054 ± 0.004		10.18 ± 0.001		0.45 ± 0.001		0.78 ± 0.002		0.93 ± 0.00	
8		8	12.25 ± 0.02		0.84 ± 0.004		12.55 ± 0.003		0.48 ± 0.000		0.19 ± 0.001		0.79 ± 0.003	
9		10	12.22 ± 0.01		0.93 ± 0.002		15.72 ± 0.002		0.72 ± 0.001		1.08 ± 0.003		0.76 ± 0.00	
10		12	12.22 ± 0.01		0.97 ± 0.002		20.41 ± 0.001		0.36 ± 0.005		1.25 ± 0.001		0.82 ± 0.004	
11	76	4	12.26 ± 0.02		0.85 ± 0.002		16.00 ± 0.000		0.84 ± 0.001		0.18 ± 0.001		0.68 ± 0.002	
12		6	12.25 ± 0.02		0.68 ± 0.002		20.47 ± 0.005	±	1.03 ± 0.005		1.74 ± 0.002		0.83 ± 0.003	
13		8	12.59 ± 0.01		1.03 ± 0.004		14.93 ± 0.004		0.77 ± 0.002		4.82 ± 0.004		1.22 ± 0.004	
14		10	12.52 ± 0.02		0.88 ± 0.004		19.67 ± 0.004		0.51 ± 0.003		5.44 ± 0.005		0.84 ± 0.00	
15		12	12.55 ± 0.02		0.98 ± 0.000		18.88 ± 0.003		0.30 ± 0.002		3.93 ± 0.001		0.86 ± 0.00	

T/C and c refer to Tailing/cement ratio and concentration, respectively. LV refers to Limited value. SS refers to suspended solids. P refers to phosphate. AN refers to ammonia nitrogen. F refers to fluoride. Texts highlighted in red are limited values in GB8978-1996 (Comprehensive Wastewater Discharge Standard), and texts highlighted in yellow are limited values from GB 14848-2017 Standard for Groundwater Quality

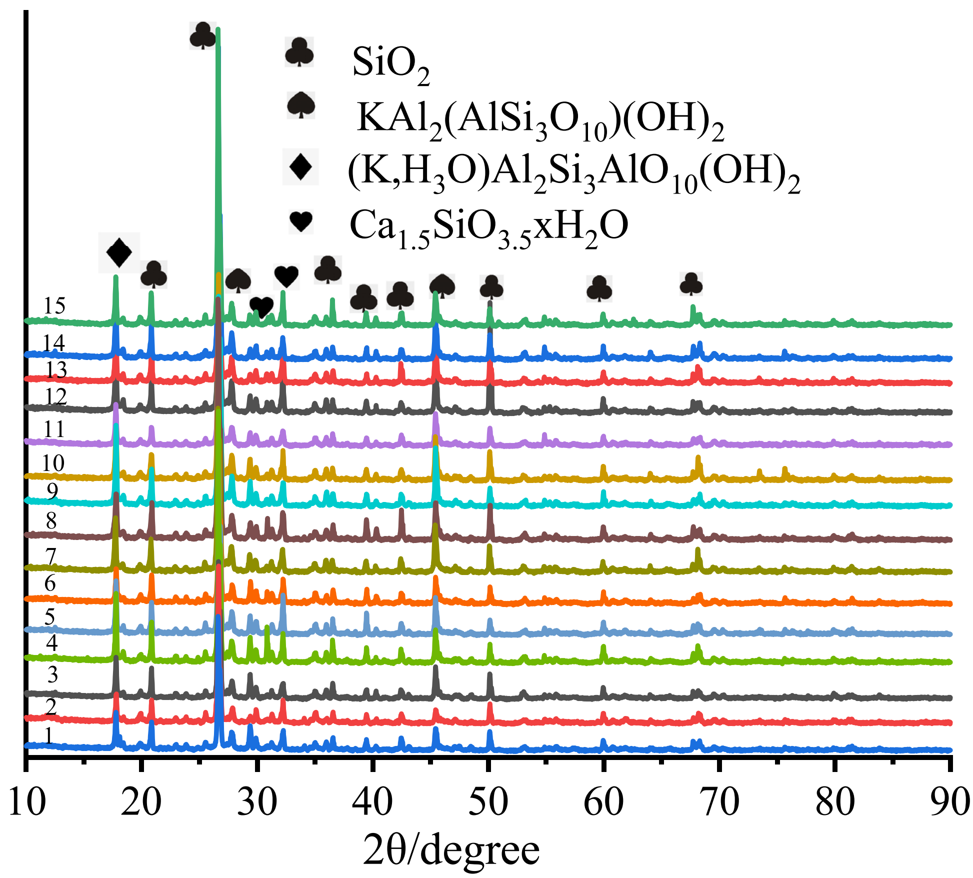
Table 8 the results of metal concentrations and leaching standard values

Element	Cu	Zn	Cd	Pb	Total Cr
Content (mg/L)	0.002 ± 0.0001	0.0059 ± 0.0002	0.001 ± 0.0001	0.0035 ± 0.0004	0.0015 ± 0.0001
Limited value (mg/L)	100	100	1	5	15
Element	Cr^{6+}	Be	Ba	Ag	Se
Content (mg/L)	0.003 ± 0.0002	0.00056 ± 0.00002	0.0012 ± 0.0002	0.0025 ± 0.0001	0.00005 ± 0.000002
Limited value (mg/L)	5	0.02	100	5	1
Element	Ni	Hg	As		
Content (mg/L)	0.0032 ± 0.0001	0.00026 ± 0.00001	0.00068 ± 0.00001		
Limited value (mg/L)	5	0.1	5		

Morphological Analysis

XRD analysis was conducted on 15 groups of tailings fill materials, and we found that the main composition was SiO_2, with the strongest diffraction peak being quartz (Fig. 6). It can be seen that silicon and aluminum elements in the tailings mainly exist as crystals, with a strong diffraction peak appearing at $2\theta = 27°$, indicating that the main mineral component is SiO_2 (Fredy et al. 2021). Broad diffraction peaks appeared near $2\theta = 30°$, 17°, and 45°, which can be attributed to hydrated calcium silicate $Ca_{1.5}SiO_{3.5}\chi H_2O$, $(K,H_3O)Al_2Si_3AlO_{10}(OH)_2$ $KAl_2(AlSi_3O_{10})(OH)_2$, respectively, indicating that the slurry was bound to the broken bonds on the surface of the tailings. Apparently, while the potassium originating from the gold mine tailings was inserted in the stratified structure, silicate substances composed of K, Si, Al, and OH were produced by a hydration reaction. The chemical composition analysis results of the tailings (see Table 1) are consistent with this, indicating that the stability of the tailings was good during the hydration reaction with the cement, and the composition of the tailings did not markedly change hydration, except for the small amount of hydration products

Fig. 6 XRD results of the filling materials



generated. The morphology of samples 1 and 5 are shown in Figs. 7 and 8. A large amount of flocculent and flaky hydration products were found in the sample (see Fig. 7a), and the fine tailings were solidified and compacted, forming a dense structure. At higher magnification, a small amount of needle-shaped ettringite were found in the sample (see Fig. 7b), and the stable hydration products (hydrated calcium aluminate) were interlaced with plate-shaped and needle-shaped substances, filling the gaps between the tailings to form a spatial network-like binding structure, thereby providing mechanical strength. Meanwhile, the good flowability of sample 1 (see Fig. 2) facilitated hydration development at the initial setting stage. However, there were less hydration products, such as portlandite, calcium silicate hydrate (C–S–H) gel, and acicular ettringite in sample 5 (see Fig. 8), which is consistent with the mechanical properties in Fig. 5. The lower strength development in sample 5 was due to a lack of hydration products, such as C–S–H gel and portlandite, and hence poor inter particle bonding. In addition, the flowability and bleeding rate of sample 5 are much less than those of sample 1 (see Figs. 2, 3), which agrees with the morphological analysis.

Consequently, the strength of the fully tailings-filled material from gold mines still originated from the hydration, condensation, and hardening of the four major minerals (C_3S、C_2S、C_3A、C_4AF), generating high calcium-silicon ratio C–S–H gel and AFt. When using ordinary Portland cement to solidify gold tailings, the tailings reacted with the alkaline activator $Ca(OH)_2$ and sulfate, where the active Al_2O_3, SiO_2, and $Ca(OH)_2$ reacted to generate a low calcium-silicon ratio C–S–H gel and AFt. This effect became more pronounced when the tailings/cement ratio was > 8, and the macroscopic manifestation was that the uniaxial compressive strength of the filling materials increased with

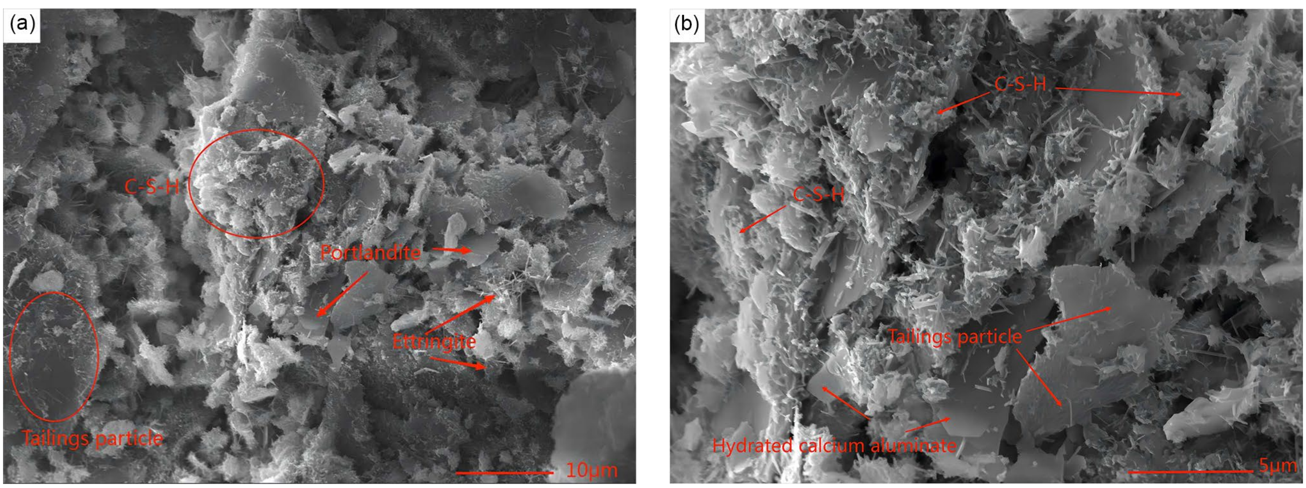


Fig. 7 SEM images of the hydration products of sample 1

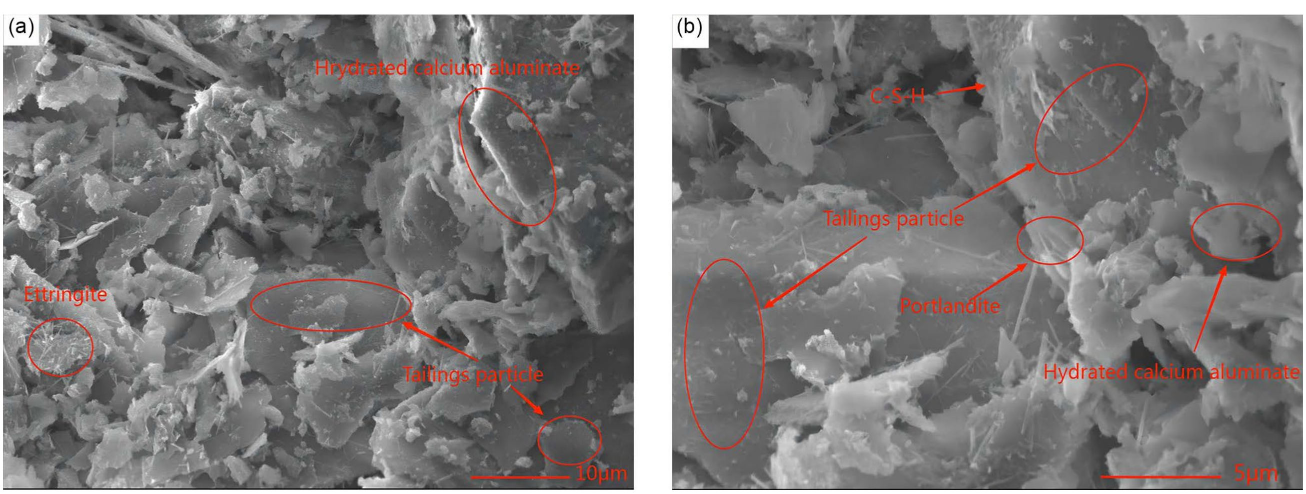


Fig. 8 SEM images of the hydration products of sample 5

the increase of the slurry concentration (Fig. 5b). It can be inferred that the low calcium-silicon ratio of the C–S–H gel and AFt and other hydration products increased the uniaxial compressive strength of the sample.

Conclusion

A one-step method was used to prepare the tailings as backfill materials. The optimal concentration and tailings/cement ratio of the tailings fill mixture were assessed and determined. It was found that under standard curing conditions, the cement hydration reaction was pivotal and the secondary hydration reaction of the tailings had less impact on the physical properties of the filling materials. The optimal axial compressive strength of the filling materials was obtained when the concentration was 68% and the tailings/cement ratio was 4:1, which is suitable for underground filling for depleted mines.

The rheological properties, coagulation-hardening properties, and bleeding rate of the fill materials deteriorated as the concentration and tailings/cement ratio increased. When the concentration was 68% and the tailings-cement ratio was 4:1, the flowability of the filling materials was 17.4 cm, the initial and final setting times were 7 h and 12 h, respectively, and the bleeding rate was 3.74%. The water quality and toxicity analysis of the leachate showed that the indicators were below the standard limit, suggesting that the filling materials can be used as backfill material for mined-out areas without causing secondary pollution.

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