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Utilization of Mine Tailings As Partial Cement Replacement

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Abstract: Depositing of mine tailings entail major economic costs and potential negative environmental impacts. Thus finding an alternative to depositing is of interest. This study focused on the use of mine tailings as a partial cement replacement, thereby preventing depositing the mine tailings. Mine tailings from two different mines Zinkgruvan (Sweden) and Nalunaq (Greenland) were both tested as 5 and 10% cement replacement. Addition of mine tailings in the fresh concrete mix affected the consistency negatively increasing the water demand, but an acceleration of hydration reactions were seen for both tailings. A lower compressive strength was obtained for mortar specimens with tailings compared to reference specimens at 7, 14 and 28 days of curing. However, both mine tailings showed indications of pozzolanic activity. This tendency was most pronounced for Zinkgruvan, indicating a higher reactivity of the oxides contributing to the pozzolanic reactions. The mine tailings has a relatively high content of toxic chemical elements, but no significant leaching of chemical elements was seen from neither the pure mine tailings nor the mortar specimens containing mine tailings. Overall, the results show that these mine tailings have potential as a partial cement replacement in cementitious mixtures.

Key words: mine tailings, partial cement replacement, compressive strength, leaching, sustainability

1. Introduction

In recent years, the pressure on the global environment has led to an increase in the demand for renewable energy source and a decrease in the utilization of coal for energy production. Coal fly ash, a byproduct from the coal energy production, has been utilized as a mineral admixture in cementitious mixtures in order to reduce the amount of CO₂ released from the cement production and further conserve raw materials in relation to the cement production [1]. However, with the conversion from the use of fossil fuels to use of renewable energy, this will not remain an opportunity. The interest in preserving the reduced emissions of CO₂ from the cement production makes the use of secondary, raw materials as a partial cement

replacement of great interest.

Mine tailings is a particulate waste material originating from mineral processing in mining activities. As the mines are constructed for different mining activities (e.g., gold and copper mines), the by-product tailings vary in quality, composition etc. depending on origin. Today tailings are landfilled and not utilized. But landfilling of tailings leads to a range of potential problems. In addition to occupation of land, costly constructions, maintenance and monitoring. Landfilling of tailings also facilitates ecological risks, such as air pollution, being a potential health risk to nearby residents or a source of pollution of the surrounding nature, and pollution of surface and underground water due to leaching of chemical elements [2].

A replacement of cement in a cementitious mixture with an admixture, such as tailings, can influence the properties of the final product. The use of tailings has not been studied widely in the literature and combined

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with a variety in the composition of the tailings due to the different origins, no general assumptions can be made on how tailings influence a cementitious mixture when utilized as a partial cement replacement.

Onuaguluchi and Eren (2012) [3] and Thomas et al. (2013) [4] studied the utilization of copper tailings in cementitious mixtures. Onuaguluchi and Eren (2012) [3] used copper tailings as an addition to cement, which increased water demand and affected the consistency negatively when added pre-dried. When pre-wetted tailings were used, this drawback was reduced and the addition of copper tailings increased the compressive strength. Thomas et al. (2013) [4] utilized copper tailings as a partial sand replacement. It was observed that up till 60% of the fine aggregates in a cementitious mixture could be replaced by the copper tailing investigated, exhibiting good strength and durability characteristics.

The aim of this work was to further evaluate the potential utilization of tailings as a partial cement replacement in a cementitious mixture and contribute to the knowledge on how different tailings behaves. Thus, two tailings from different types of mines (gold and zinc, copper and lead) are included in this work. In addition to the influence on cementitious mixtures, such use also calls for an evaluation of the leachability of toxic elements to prevent spreading of contamination in regards to the later demolition, depositing at landfills or possible recycling.

1.1 Mine Tailings as A Construction Material

Mine tailings are not used currently in concrete, and there are no standard available for such use. The limits presented in UNI EN 450-1 [5], the current European standard for use of fly ash as a mineral admixture in cementitious mixtures, will be used for evaluation in this work.

Tailings in a cementitious mixture could contribute either as a pozzolan, which can replace parts of the cement or as an inert filler, which can replace sand and/or fine aggregates. The pozzolanicity is governed

by the chemical composition of primary oxides and the total percentage of silicon dioxide, aluminium oxide, and iron oxide should be at least 70% [5, 6]. Fillers are defined as particles with a maximum particle size of 250 μm [7, 8]. Fillers are considered inert, but not without influence. Fillers are able to fill the intergranular voids in between the cement grains of a cementitious mixture, thus decrease the porosity and increase the compressive strength, known as the filler effect [8, 9]. An inert filler may further be able to provide a heterogeneous nucleation effect, acting as nucleation sites for the hydrates in the cement, accelerating the hydration reactions and thus improving the compressive strength [8, 10, 11]. The ability of the tailings to act as nucleation sites depends on the fineness of the particles, the amount of mineral admixture and the affinity of the mine tailing to cement hydrates, related to the origin of the mine tailing [10, 12]. The heterogeneous nucleation effect develops at an early age and is maintained over time [10]. Generally, a replacement of cement with an inert filler leads to a decrease in the compressive strength with an increase in the replacement rate [10].

2. Material and Methods

2.1 Materials and Characterization

Two different types of mine tailings were investigated in this work:

- Zinkgruvan (Z): Mine tailing from zinc, copper and lead mine in Sweden. The deposit is a part of the Proterozoic aged Berslagen greenstone belt, which hosts massive Zn, Pb, Cu and Ag sulphides and banded iron formations in volcano-sedimentary complexes [13]. The tailings consist mainly of quartz, feldspar and calcite [14].
- Nalunaq (N): Mine tailing from the gold mine Nalunaq in the southern part of Greenland. The deposit is classified as an orogenic-type gold mineralization and is hosted in amphibolite facies meta volcanic rocks [15].

The particle size distribution, mean particle size and specific surface area was found by laser diffraction. The pH was measured with a pH electrode and the conductivity with an electrical conductivity meter in a 1:2.5 solid to liquid suspension in distilled water. The solubility was measured by preparing a 1:5 solid to liquid ratio suspension with distilled water. The suspension was shaken for 1 min and decanted. Additional 500 mL of distilled water was added. This procedure was repeated three times before the suspension was filtered and the sample dried and weighed. Loss on ignition (LoI) was measured in accordance with UNI EN 196-2 [16] at 950°C for 5 min with lid followed by 10 min without lid. The mineral content was analyzed by X-ray diffractions (XRD) measured by PanAlytical X-ray diffractometer. The chemical composition was determined by X-ray fluorescence (XRF). The morphology of the mine tailing particles was evaluated with a Scanning Electron Microscope (SEM).

2.2 Material Testing

The cement used was Portland Basic Cement (CEM II). Mortars were mixed according to UNI EN 196-1 [17]. Mortars with 0%, 5% and 10% replacements of cement with mine tailings were mixed. The notations in the following sections are as follows:

- REF is a reference mixture without any replacement.
- 5 Z and 5 N are 5 % wt. replacements of cement with tailings from Zinkgruvan (Z) and Nalunaq (N).
- 10 Z and 5 N are 10 % wt. replacements of cement with tailings from Zinkgruvan (Z) and Nalunaq (N).

Mine tailings were dried at 105°C for 24 h to a water content of 0% before mixed into the cementitious mixtures.

The consistency was measured on fresh mortar according to UNI EN 1015-3 [18] and the setting

behaviour was measured by Vicatronic according to UNI 196-3 [19].

Mortar specimens (40×40×160 mm) were cast for all mixtures (0%, 5% and 10% replacements with tailings) in accordance with UNI EN 196-1 [17]. The specimens were de-moulded after 24 hours and submerged in water to cure for 7, 14 and 28 days. The compressive strength of the mortar specimens was tested according to UNI EN 196-1 [17].

The leaching of As, Cd, Cu, Ni, Pb and Zn from mortar specimens containing mine tailings was measured by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) after preparation as follows: the mortar specimens were pulverised using a hammer and dried at 105°C. A suspension with 20 g of the pulverised mortar and 40 mL of distilled water was prepared and placed at an agitating table for 23 hours. The solution was left to settle for 15 min and afterwards filtered. The leaching of chemical elements was measured in the filtrate in duplicate.

3. Results and Discussion

3.1 Characteristics of Investigated Mine Tailings

The cumulative particle size distribution is displayed in **Fig** for Zinkgruvan and Nalunaq, with CEM II for comparison. According to UNI EN 450-1 [5], 40% of the particles must be retained when sieved on a 45 µm sieve. Neither Zinkgruvan nor Nalunaq comply with this limit and concerning the particle size distribution neither of the tailings are expected to contribute significantly to the filler effect. This is substantiated by Moosberg-Bustnes et al. (2004) [8] describing that only particle smaller than 125 µm contributes to the filler effect. 70% of Nalunaq and only 27% of Zinkgruvan conforms with this limit.

The physical properties and the chemical composition of the tailings are given in Table 1, together with values obtained by a literary review on coal fly ash used today in the cement production [20-30], for comparison. From the chemical composition, it is seen that the total percentage of the

primary oxides silicon dioxide, aluminium oxide, and iron oxide does not fully meet the limit set by UNI 450-1 [5] and ASTM International C168-15 [6] of 70%. However, Nalunaq reached approximately 60% primary oxides and are, therefore, expected to

contribute more to the pozzolanic reaction compared to Zingruvan, which reach approximate 50% primary oxides. Though, neither can be categorized as a pozzolan.

Table 1 Chemical and physical characteristics of investigated raw material, \pm defines the standard deviation, Coal fly ash (CFA) are used for comparison.

	Zinkgruvan	Nalunaq	Coal fly ash interval from literary review [20–30]
Chemical composition (%)			
SiO ₂	35.5	38.7	26.0–60.3
Al ₂ O ₃	6.8	10.0	10.6–28.1
Fe ₂ O ₃	6.4	11.2	4.0–15.71
Σ (primary oxides)	48.7	59.9	40.6–100
CaO	13.6	13.7	2.0–16.6
MgO	4.0	5.0	0.89–2.8
K ₂ O	4.1	0.5	0.2–4.43
Na ₂ O	< 0.7	< 0.7	0.2–4.3
TiO ₂	0.1	0.8	0.5–1.2
SO ₃	2.0	0.7	0.3–6.2
LoI	5.1 \pm 0.5	1.4 \pm 0.0	0.4–10.4
Physical properties			
Mean particle size (μ m)	183.1	90.7	3.74–31.3
Specific surface area (m ² /kg)	67	131	0.2–0.7
pH	7.7 \pm 0.0	8.2 \pm 0.1	10.4–13.5
Conductivity (mS cm ⁻¹)	1.2 \pm 0.1	0.6 \pm 0.0	
Solubility (%)	0.92 \pm 0.2	0.75 \pm 0.2	

The mineralogy was analyzed qualitatively and “x” marks a detected mineral in

Table . Both tailings contained quartz, microcline, amphibole and plagioclase feldspar. Nalunaq further contains kaolinite and Zinkgruvan further contains calcite and dolomite. Both types of tailings are of volcanic origin. This can lead to a further contribution to the pozzolanic activity from the amorphous material (detected, not quantified) contained in the tailings since volcanic rocks are categorised as a natural pozzolan [31].

The contribution to the pozzolanic activity is further dependent on the reactivity of the admixture [31]. The conductivity, shown in

Table , gives an indication of the amount of soluble ions present in the mineral admixture and thus the

reactivity. Further, Davison et al. (1974) [32] and Lui et al. (2004) [33] described a

Table 2 Mineralogy of mine tailings determined by XRD diffractiogram.

	Zinkgruvan	Nalunaq
Quartz (SiO ₂)	x	x
Microcline (KAlSi ₃ O ₈)	x	x
Amphibole (Ca ₂ (Mg,Fe,Al) ₅ (Si,Al) ₈ O ₂₂ (OH) ₂)	x	x
Kaolinite (Al ₂ Si ₂ O ₅ (OH) ₄)		x
Calcite (CaCO ₃)	x	
Plagioclase feldspar (CaAl ₂ Si ₂ O ₈)	x	x
Dolomite (MgCa (CO ₃) ₂)	x	

decreasing reactivity, when the particle size increases due to the lower specific surface area. As displayed in Table both tailings have significantly higher specific surface area compared to coal fly ash, which is

substantiated by the SEM analysis, see Fig. 1, where both tailings are seen to consist of large, flaky particles. Zinkgruvan has both a higher specific surface area and a higher conductivity compared to Nalunaq, and thus the Zinkgruvan mine tailing is expected to be most reactive of the two.

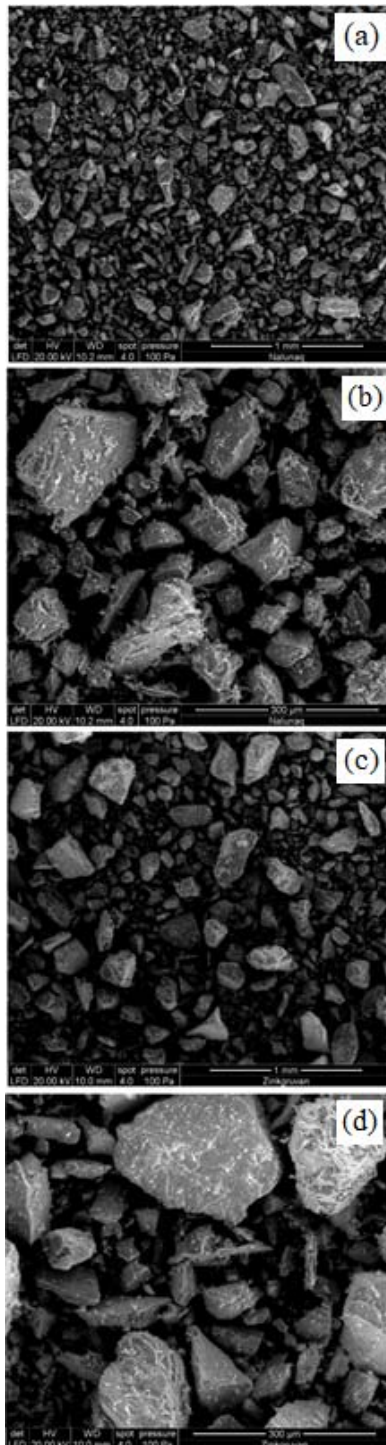


Fig. 1 SEM analysis of Nalunaq magnified 50 times (a), Nalunaq magnified 200 times (b), Zinkgruvan magnified 50 times (c) and Zinkgruvan magnified 200 times (d).

3.2 Consistency

Consistency is one of the two components constituting the workability of a cementitious mixture; consistency and stability, both accounting for the ease of which a freshly mixed cementitious mixture can be mixed, placed, compacted and finished [34]. Consistency is a simple index for the flowability, thus describing the ease of the flow [20].

Normalized flow values for fresh mortar with 0%, 5% and 10% cement replacements by tailings are displayed in Fig. 2. The addition of tailings affected the consistency negatively increasing the water demand, which corresponds to observations made by Onuaguluchi and Eren (2012) [3]. This impact may lead to difficulties during placement and compaction, which could lead to a decrease of the durability [20].

3.3 Setting Behaviour

Setting refers to the solidification of the cement paste in a cementitious mixture. Initial setting time refers to the point when the dormant stage of the cement paste has ended and the cement paste has become unworkable [20]. Final setting time marks when the paste has become completely rigid and the development of compressive strength begins [20]. Addition of a filler can shorten the setting time and accelerate the setting by providing heterogeneous nucleation centres in the initial paste solution [35]. However, addition of a filler can also decrease the hydration rate leading to a setting delay, even though this effect is mainly considered fly ash and silica fume [36]. This setting delay was also observed when copper tailings are used as an additive in a cementitious mixture by Onuaguluchi and Eren (2012) [3].

The setting process of fresh mortar with 0%, 5% and 10% cement replacements with tailings is shown graphically in Fig. , whereas initial and final setting times are given in

Table . In general, replacements with the mine tailings prolonged the dormant stage with an increase in the replacement rate, postponing the initial setting time. However, as seen from

Table , the time from initial setting time to final setting time decreased with cement replacement with tailings, which was most profound for Nalunaq compared to Zinkgruvan. These results indicate an acceleration of the hydration reactions when the tailings are used as a cement replacement in a cementitious mixture, attributed to the heterogeneous nucleation effect. This would cause Nalunaq to have the largest contribution to the heterogeneous nucleation effect, due to the smaller particle size (see Fig) and replacements with 10%, since the probability for nucleation sites to be near to cement particles increases with the amount of foreign particles [10].

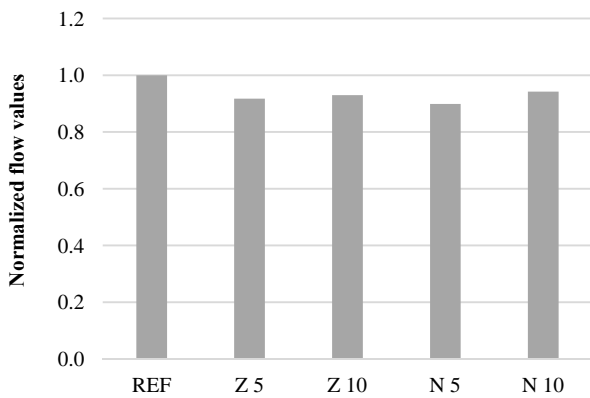


Fig. 2 Normalized flow values for fresh mortar with 0%, 5% and 10% cement replacements with mine tailings.

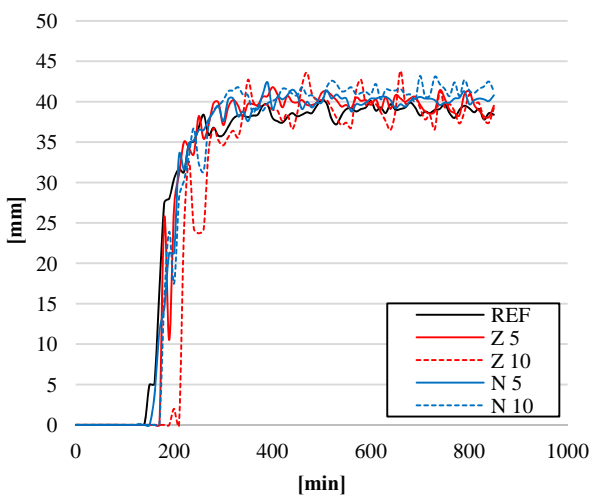


Fig. 3 Setting process for fresh mortar with 0%, 5% and 10% cement replacements with mine tailings.

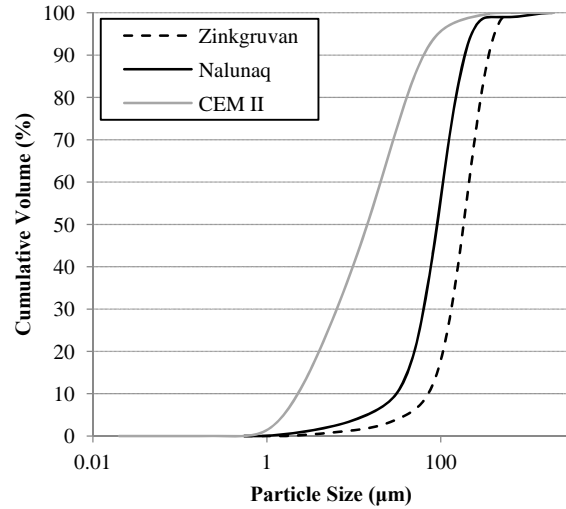


Fig. 4 Cumulative particle size distribution of test materials.

Table 3 Initial and final setting time for fresh mortar with 0%, 5% and 10% cement replacements with mine tailings.

	Initial setting time [min]	Final setting time [min]
REF	150	500
Z5	150	400
Z10	220	470
N5	170	390
N10	180	420

3.4 Compressive Strength

Fig. 5 displays the compressive strength at 7, 14 and 17 days of curing of mortar specimens with 0%, 5% and 10% cement replacements with tailings. All specimens containing tailings had a lower 7 days compressive strength compared to the reference. The difference became more pronounced at 14 and 28 days of curing, except for 5% Zinkgruvan at 28 days, which obtained 95% of the compressive strength of the reference.

For 10% Zinkgruvan and 10% Nalunaq, the compressive strength decreased slightly from 7 to 14 days. This is a reflection of the consistency of fresh mortar decreasing with an increase in the replacement rate, making the fresh mortar dry and difficult to place and compact at high replacement rates.

5% Zinkgruvan had a lower compressive strength at both 7 and 14 days of curing compared to 5% Nalunaq. This increase for 5% Nalunaq could be due to both the filler effect and the heterogeneous nucleation effect, where, for both effects, Nalunaq has been discussed to have the largest possible contributions. But 10% Zinkgruvan and 10% Nalunaq obtained the same compressive strength at every curing duration indicating no significant differences between the two tailings regarding contributions to either effects. Further, a decrease in the compressive strength by increasing the replacement percentage was seen for both Zinkgruvan and Nalunaq. This could indicate that no heterogeneous nucleation takes place and no contribution to the filler effect is obtained or it could merely be a result of the reduced cement content.

The compressive strength of all specimens is seen to increase from 14 days to 28 days of curing. Specimens containing Zinkgruvan obtained the highest compressive strength of those containing tailings at both 5 and 10% replacement at 28 days of curing. This indicates the highest contribution from the pozzolanic activity to the compressive strength for specimens containing Zinkgruvan. Since Nalunaq was determined to contain the largest amount of primary oxides contributing to the pozzolanic reactions, the oxides contained in Zinkgruvan could be more reactive in order to have a larger contribution to the compressive strength, corresponding with the results for the specific surface area and conductivity.

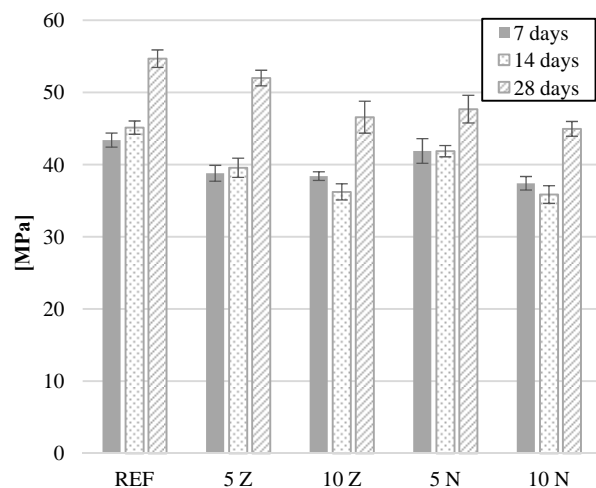


Fig. 5 Compressive strength of mortar samples with 0%, 5% and 10% cement replacements with mine tailings.

The reaction between the tailings and the calcium content in the cement might lead to some limitations in the use, as this may influence the mechanical properties of a mortar specimen when used as a partial cement replacement [2]. The impact of the reactions between calcium and the tailings may also be reflected in Fig. 5, displayed by a reduction of the compressive strength, but are not separately identified.

It should be noted, that in this study only indications of pozzolanic activity have been observed. In order to determine whether the tailings have a contribution the compressive strength through pozzolanic activity, compressive test of mortar specimens at 90 days of curing are needed [5].

3.5 Leaching Characteristics

Table 4 shows the initial content of selected chemical elements in the tailings and the limit for the content of chemical elements of residues and soils for building and construction according to note number 1662/2010 [37]. The initial content of the selected chemical elements As, Cd, Cu, Ni, Pb and Zn does not comply with this set of limits.

The leaching characteristics of pure tailings and specimens containing 5 and 10% replacements are shown in Table 5. The concentrations of chemical elements in the leachate from the mortar specimens containing 5 and 10% replacements are significantly

lower than from the initial content of the selected chemical elements in the tailings. Ahmari and Zhang 2013 [2] described a possible stabilisation of deposited

Table 4 Initial content of selected elements in mine tailings (mg/kg±standard deviation) [38]. Limit according to the Danish Environmental Protection Agency Miljøstyrelsen (2010) [37].

	Limit	Zinkgruvan	Nalunaq
As	20	21±2	122±35
Cd	0.5	11±0.2	2.8±0.9
Cu	500	372±28	105±28
Ni	30	24±3	48±13
Pb	40	3,700±233	59±22
Zn	500	7,331±322	45±22

Table 5 Leaching of selected elements in mortar specimens containing 5 and 10% cement replacement respectively (mg/kg±standard deviation).

	REF	Z5	Z10	N5	N10
As	0.0±0.0	9.7±8.5	11.6±2.1	0.0±0.0	11.9±0.5
Cd	0.0 ± 0.0	0.9 ±0.7	0.0 ±2.2	0.0±0.9	0.0 ±0.0
Cu	0.0 ± 0.0	1.4 ±0.4	4.5 ±0.4	2.0±1.6	3.5 ±0.5
Ni	0.0±0.0	0.0±0.4	0.8±0.3	0.0±0.9	1.1±0.5
Pb	0.0±0.0	12.4±11.7	2.5±0.5	0.0±0.0	0.5±0.9
Zn	0.0±0.0	3.6±6.1	0.3±0.5	0.0±0.0	0.5±0.7

tailings with cement, due to tailings reacting with the calcium content in cement. The tailings are thus chemically stabilised by a hardened surface, which isolates the underlying tailings from the environment reducing the amount of chemical elements leaching, corresponding well with the obtained results in Table 5. Also, the high pH in mortar can cause precipitation of heavy metals and lowers the leachable fraction.

4. Conclusion

The use of two different mine tailings as a partial cement replacement has been studied in this paper. The major conclusions are:

- The addition of mine tailings affected the consistency of the fresh mortar negatively increasing the water demand by causing the mortar to be dry and difficult to place and compact in the moulds.

- Both mine tailings facilitated an acceleration of the hydration reactions when utilized as a cement replacement.
- Specimens with partial cement replacement with mine tailings from Nalunaq and Zinkgruvan obtain a lower compressive strength compared to the reference specimen without mine tailings.
- Zinkgruvan contributed most to the compressive strength at 28 days of curing, probably due to higher reactivity of the minerals contributing to the pozzolanic activity, compared to Nalunaq.
- The initial content of specific chemical elements in the mine tailings exceed the limits allowed in residues and soils for building and construction according to note number 1662/2010 [37].
- Significant leaching of toxic chemical elements from the mortars with mine tailing was seen. This issue needs to be discussed both in regards to the emission to the surrounding environment during use and at end of service life.

In conclusion, both Zinkgruvan and Nalunaq could be utilized as a partial cement replacement. However, further studies are needed, especially on the pozzolanic activity of the mine tailings and the issue on leaching needs to be addressed.

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