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Geopolymerization of Mining Tailings as an Alternative for Its Use in the Construction Industry

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ABSTRACT

The use of mining tailings-based geopolymer binder as a substitute for cement can be an interesting alternative to the problem of final disposing of large volumes of them. Indeed, mining tailings have a high silica content, which makes them suitable for geopolymerization. The tailings samples contain Cu, Pb, Zn, As, Sb, Cd and Bi, mainly. X-ray diffraction results show that the tailings are composed of quartz, SiO_2 (81%). Geopolymerized tailing samples (GTPS) were obtained by varying the ratio of SiO_2/Al_2O_3 and SiO_2/NaOH. The compressive strength of the GTPS varied between 2 MPa and 8 MPa. The electrical conductivity in the leaching tests varies between 13.91–16.11 mS/cm^2 indicating that ions are present in the solution. However, the pH = 10.5 indicates that acidity is not being generated. The element that mostly leaches is iron. The highest percentage of leaching is observed for cadmium (69%) and chromium (93%). The concentrations of elements are under the maximum permissible limits for water according to the Peruvian legislation. However, mass transfer leach tests and immersion leach tests must be realized.

INTRODUCTION

Mining is an important industry in the economy of many countries. For mining operations to develop, the movement of large volumes of material is necessary. In this sense, this industry generates a variety of solid and semi-solid waste, with tailings being one of the largest volume products generated [1]. In fact, the mining industry generates a huge amount of mine tailings from the excavation of minerals in the form of waste rock from mineral processing [2]. In this sense, it is necessary to implement an adequate management system for the waste generated [3,4].

In recent years, the mining industry around the world has focused its efforts on minimizing this waste, either by reusing it in its operations with the cut-and-fill method [5,6] or looking for alternative uses in society [7]. Another problem with tailings is related to the handling of large volumes that must be disposed of safely [8–10]. One way to reduce or minimize the volume of this waste is to use it as raw material for the production of inputs for other industries [11], such as construction [10, 12–16]. The literature has shown that it is possible to use copper tailings [2,17,18], gold [18–22] and iron [23–30] mixed with geopolymers [18,23,25] as partial Cement substitute for concrete or making construction bricks. In this sense, the use of geopolymerized mine tailings is a new trend of great relevance in the mining industry and especially useful for the reuse of tailings [31–33, 34]. In this article, the preliminary results of obtaining geopolymerized cement from mine tailings are presented.

MATERIALS AND METHODS

Collection of Mining Tailings

The copper-gold mining tailing sample came from a mining operation in northern Peru. The sample containing 25% solids was dried at a temperature around 50°C for 5 days [35]. This tailing was pretreated in order to eliminate the percentage of sulfur [36].

Tailing Characterization

The Inductively Couple Plasma (ICP) analytical method was used to determine the heavy metal concentrations in each sample of liabilities.

Mineralogical Tests

To obtain the mineralogical composition, X-ray diffraction (XRD) was used. The XRD analysis was performed with the DRX Bruker equipment model D8 Discover with copper radiation (Cu Kα = 0.15418 nm), current of 40 mA and acceleration voltage of 40 kV with a Lynxeye detector with energy selectivity.

The analysis was performed in a range of angles (2θ) from 5 to 80 degrees in steps of 0.02 degrees considering the time per step 1 second. To calculate the composition of the crystalline phases and the amorphous part, the Reference Intensity Ratio (RIR) method was applied [37].

X-Ray Flourescence

One gram of material was selected for each sample, X-ray fluorescence analyses. X-ray flourescence analyzes were performed with the Bruker S8 TIGER wavelength dispersive with opening angle of 0.23° and 0.46°. In addition to analyzer crystals: PET, LIF (200), LIF (220) and XS-55, and two types of detectors: scintillation counter and proportional flow. The evaluation model was for oxides and the measurement time was ~17 minutes.

Chemical Speciation

The concentrations of heavy metals in the tailing were determined using Inductive Couple Plasma (ICP) method. To determine the concentrations of metals in the leaching tests, the Atomic Absorption Spectroscopy (AAS) technique was used [38].

Acid Base Accounting (ABA) analysis

The acid mine drainage (DAM) prediction analyzes were carried out following the procedure proposed by Skousen [39]. Then, a sample of 2.0 g of pulverized sample were placed into 250 mL conical flask and 90 mL of distilled water was added. At the beginning of the test, between 1–3 ml of standardized 1,0 N HCl was added. Then the flask was placed on a shaking apparatus. After 2 hours, a second acid quantity was added. After 24 hours, distilled water was added to bring a volume of 125 mL approximately. Then, the pH was registered before start the neutralization up to 8.3 by adding a standardized solution of 0,5 N or 0,1 N NaOH.

Radiometric Analysis

The radiometric analysis was carried out using gamma-ray spectrometry with a 3x3" NaI(Tl) scintillation detector to determine the activity concentrations (AC) of natural radionuclides. The tailing samples were placed in cylindrical hermetic containers to prevent radon leakage and stored for a period of 28 days to reach secular equilibrium [40].

Quantifications were carried out by the efficiency transfer method using the software ETNA and the standard reference materials IAEA-412 and IAEA-465 with known activities of 40K, 226Ra and 232Th. The 1460 keV, 1764.5 keV and 2614.7 keV gamma lines were used for estimating ACs of 40K, 214Bi(226Ra), and 208Tl(232Th), respectively [41,42].

Geopolymerization Process

For geopolymerization to be carried out, it will be necessary to adjust the molar ratio of Si/Al and Si/Na. In this case, the Si/Al ratio was fixed at 1 and 5 by adding aluminum oxide to the mixture. In the case of the Si/Na ratio, this was set between 0.5 and 2.5. The concentration of sodium hydroxide used in the alkaline solution was set from 9 to 13 molar.

Six geopolymer recipes were prepared by varying the ratios of Si/Al and Si/Na. All recipes were molded in cubic molds of 5cm based on the ASTM C109 standard [43]. After molding, the geopolymers were cured at ambient temperature for 15 days. After this period, the geopolymers were further cured in a furnace at a temperature of 75°C for 7 weeks.

Geomechanical Tests

The standard method provides a means of determining the compressive strength of hydraulic cement and other mortars and results may be used to determine compliance with specifications [43].

Leaching Tests

The tailings samples were mixed with the geopolymers to obtain cubic bricks of five centimeters in measurement. These bricks were subjected to leaching tests in order to identify the metals that could leach after obtaining the geopolymerized cement [44].

In addition, this will allow evaluating if the geopolymerized cement will have an acidity-generating potential, thus complementing the ABA tests.

RESULTS AND DISCUSSION

Tailing Characterization

Mineralogical Tests

The results of X- ray fluorescence and XRD analysis of the mine tailing sample R5 are presented in Table 1 and Figure 1, respectively. The results show that the sample is predominantly composed of Quartz (SiO_2), with varying concentrations of other minerals such as Muscovite ($KAl_2(AlSi_3O_{11})(OH)_2$), Wollastonite ($CaSiO_3$) and Caolinite ($(Al_2Si_2O_5(OH)_4)$.The mineral phases identified in the sample are of particular interest due to the aluminosilicates compounds for making geopolymer cement based on tailings.

Chemical Analysis

The results of the chemical analysis of the tailing sample (R5) using ICP-MS are shown in Table 2. The results show that the sample contains varying concentrations of several elements, including Cu, Cr, Mn, Pb and Zn.

Table 1. Mineral composition of sample R5

Crystalline phase	Formula	Concentration (%)
Quartz	SiO_2	81
Muscovite	$KAl_2(AlSi_3O_{11})(OH)_2$	2.7
Wollastonite	$CaSiO_3$	1.5
Caolinite	$Al_2Si_2O_5(OH)_4$	1.0
Hematite	Fe_2O_3	0.7
Rutile	TiO_2	0.4
Trace elements	Amorphous	12.7

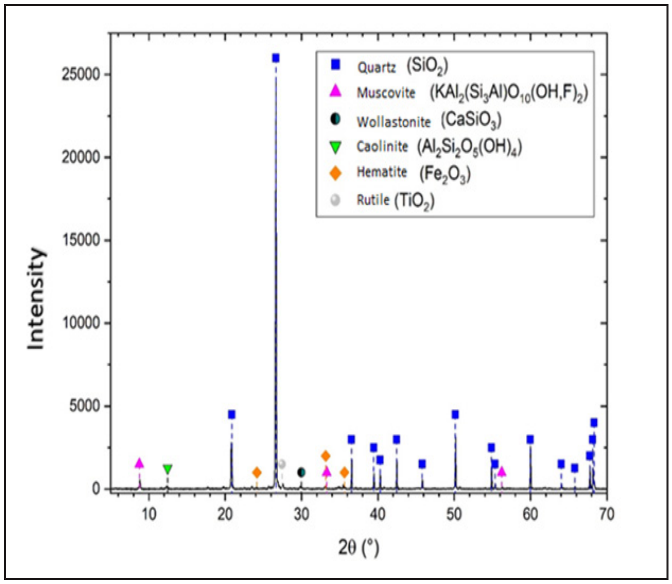

Figure 1. XRD spectrum of tailing sample (R5)

Radiometric Analysis

Activity concentrations of 226Ra, 232Th and 40K measured in the tailing sample together with the statistical error are given in Table 3.

Geopolymerization Process

Geopolymerized tailing samples with different proportions of SiO_2/Al_2O_3 and $SiO_2/NaOH$ varying from 1 to 5; and 1 to 2 respectively were obtained [45]. The samples were

Table 3. Activity concentration ($Bq.Kg^{-1}$) of ^{226}Ra, ^{232}Th and ^{40}K in tailing samples

Sample	AC_{40_K}	$AC_{226_{Ra}}$	$AC_{232_{Th}}$
R5	752.4 ± 35.7	9.9 ± 0.5	11.9 ± 0.6

Table 2. Chemical composition obtained by ICP-MS[1]

Ag ppm	Al %	As ppm	Ba ppm	Be ppm	Bi ppm	Ca %	Cd ppm	Co ppm	Cr ppm
0.3	2.99	6	166	<0.5	<5	0.43	1	6	323
Cu ppm	**Fe** %	**Ga** ppm	**K** %	**La** ppm	**Li** ppm	**Mg** %	**Mn** ppm	**Mo** ppm	**Na** %
448.4	6.03	19	1.61	4.5	5	0.29	259	16	0.2
Nb ppm	**Ni** ppm	**P** %	**Pb** ppm	**S** %	**Sb** ppm	**Sc** ppm	**Sn** ppm	**Sr** ppm	**Ti** %
1	7	0.04	24	1.08	5	2.7	<10	69.3	0.08
Tl ppm	**V** ppm	**W** ppm	**Y** ppm	**Zn** ppm	**Zr** ppm	**S_{total}** %	**S** %	**SO_4^{2-}** %	**Hg** ppm
<2	56	<10	2.2	96.7	10.7	1.1	0.87	0.23	0.01

[1]Analysis performed at SGS laboratory

Table 4. Results of geomechanical tests of geopolymerized tailings samples

Sample	SiO_2/Al_2O_3	SiO_2/NaOH	Simple Compressive strength (MPa)
RR5-R1	0.95	1.52	6.40
RR5-R2	1.98	1.46	3.87
RR5-R3	3.07	1.50	5.19
RR5-R4	4.29	1.47	5.23
RR5-R5	5.54	1.49	4.06
RR5-R6	2.90	0.47	2.00
RR5-R7	3.13	0.96	2.50
RR5-R8	3.17	1.45	4.94
RR5-R9	3.12	1.90	7.29

obtained by mixing the respective reagents and the pretreated R5 tailing. The proportions used are presented in Table 4.

Geomechanical Tests

The results of the simple resistance test of geopolymer specimens are shown in Figure 2 and Table 4. The results indicate that the compressive strength of geopolymer samples varies from 2 MPa to 8 MPa. The Greater resistance was achieved in the samples with a ratio SiO_2/Al_2O_3 of 3.12 and a SiO_2/NaOH ratio of 1.90, while the lowest resistance was observed in the samples with a SiO_2/Al_2O_3 of 2.90 and a SiO_2/NaOH ratio of 0.47 [45].

According to the Peruvian technical standard [46], type 1 and type 2 bricks must meet the minimum compression resistance of 4.9 and 6.0 MPa. While the value of 2.0 MPa is the minimum required for blocks used in non-load-bearing wall construction.

Acid Base Accounting (ABA) Analysis

The results of the ABA test are summarized in Table 5. According to the Net Neutralization Potential (NNP) classification criteria [47] and the Neutralization Potential/Acidity Potential (NP/AP) classification criteria [48], only the R5 tailings sample and the pretreated tailings sample (RR5) are potential generators. mine acid drain (DAM).

Leaching Tests

For leaching tests, only geopolymerized samples that presented a simple resistance test factor greater than or equal to 4.0 were taken into account. The results of the leaching tests are presented in Figure 3 and Table 6. The electrical conductivity in the leaching tests varies between 13.91–16.11 mS/cm^2 indicating that ions are present in the solution.

The pH of the resulting solution is in the basic range for all geopolymerized samples. The concentrations of leached elements are still below the maximum permissible limits for water according to Peruvian legislation [49]. The elements that leach in the greatest quantity are cadmium and chromium with 69% and 93% respectively.

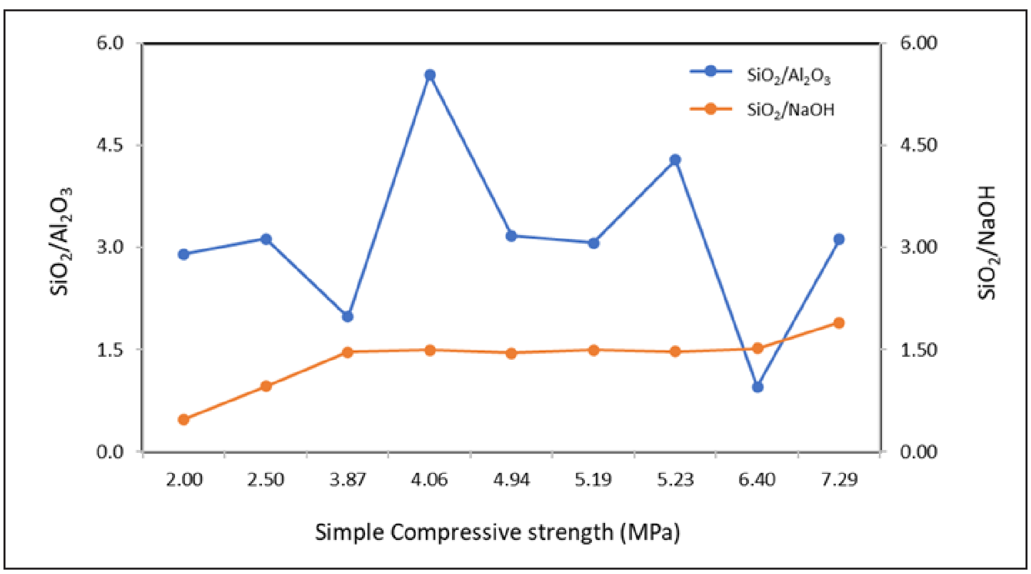

Figure 2. Variation of simple comprehension tests of geopolymerized samples

Table 5. Results of geomechanical tests of geopolymerized tailings samples

Sample	Acidity potential (AP)	Neutralization potential (NP)	Net neutralization potential (NNP)	NP/AP
R5	27.03	5.47	-21.56	0.20
RR5	17.19	3.94	-13.24	0.23
RR5-R1	4.55	107.24	102.69	23.57
RR5-R2	5.76	87.60	81.84	15.20
RR5-R3	6.32	79.61	73.29	12.60
RR5-R5	6.82	86.68	79.85	12.70
RR5-R8	6.47	86.24	79.76	13.32
RR5-R9	7.28	81.19	73.91	11.16

Table 6. Results of the leaching tests of the geopolymerized samples

Sample	Condusctivity (uS.cm^{-2})	pH	mg/L					
			Cu	Zn	Fe	Cr	Cd	Pb
RR5	0.19	6.22	0.450	0.010	0.572	0.160	0.004	0.238
RR5-R1	14.86	10.54	0.159	0.079	4.562	0.251	0.039	0.398
RR5-R2	14.41	10.53	0.103	0.056	5.352	0.374	0.041	0.450
RR5-R3	16.11	10.85	0.043	0.030	2.298	0.229	0.052	0.355
RR5-R5	15.03	10.72	0.070	0.019	2.909	0.268	0.046	0.385
RR5-R8	13.91	10.65	0.184	0.053	7.780	0.201	0.043	0.303
RR5-R9	14.60	10.72	0.151	0.093	8.780	0.159	0.049	0.210

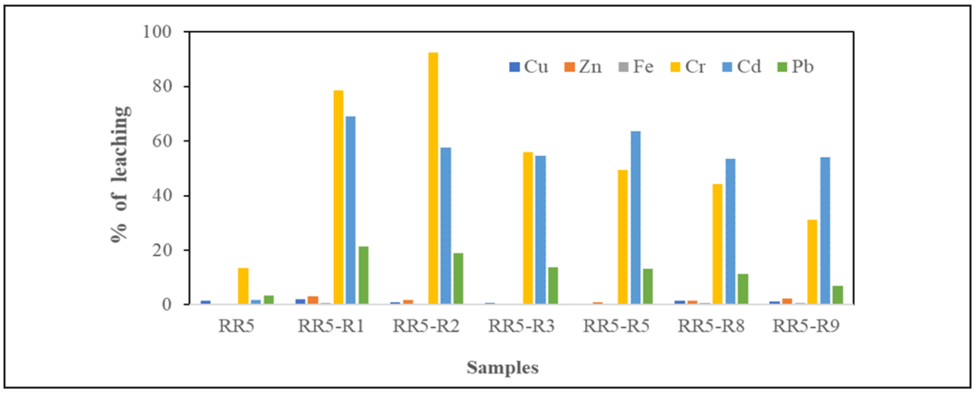

Figure 3. Comparison of metal leaching percentages in geopolymerized samples

Table 7. Metal leaching percentages in the geopolymerized samples

Sample	% Cu	% Zn	% Fe	% Cr	% Cd	% Pb
RR5	1.5	0.1	0.2	13.5	1.8	3.4
RR5-R1	2.0	3.1	0.5	78.7	68.9	21.3
RR5-R2	1.0	1.7	0.5	92.6	57.6	19.0
RR5-R3	0.6	0.5	0.2	55.9	54.5	13.8
RR5-R5	0.4	0.8	0.2	49.3	63.5	13.1
RR5-R8	1.6	1.5	0.7	44.2	53.4	11.4
RR5-R9	1.2	2.3	0.7	31.1	54.1	7.0

CONCLUSION

The proportions of SiO_2/Al_2O_3 and SiO_2/NaOH are a key factor for the proper formulation of geopolymerized samples. Results indicate that it is possible to use geopolymerized tailings as raw material for making bricks. Geomechanical tests indicate that it is possible to manufacture type I and type II bricks since the compressive strength results ranged between 2 MPa and 8 MPa. The electrical conductivity values (13.91–16.11 mS/cm^2) of the leachates from the leaching tests of the geopolymerized samples show a slight leaching of the material. Leaching tests show that bricks do not generate acidic solutions (pH ≥ 10.5). The concentrations of the leached elements (Cu, Cd, Zn, Fe, Cr and Pb) of the geopolymerized samples are below the maximum permissible limits despite the fact that the highest leaching percentages are of cadmium (69%) and chromium (93%). These results show that geopolymeric cement made from tailings can be used as a good material for infrastructure with many advantages over conventional cement, such as mechanical resistance, low levels of leached heavy metals, low concentration levels of natural radioactivity with a low degree of radiation contamination.

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Competing interest

The authors declare that they have no conflict of interest in the publication.

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Geosensing for Exploration—New Technology for Underground Directional Drilling

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ABSTRACT

Geosensing is used in oilfield drilling in conjunction with geophysical tools to support characterization of rock properties, geology, and reservoir conditions. This technology is now available to the mining industry and has been successfully applied to underground in-seam directional drilling in Australian coal mines for geologic exploration and methane drainage. The system utilizes a range of drill rig and wellhead mounted sensors which, combined with analytical software, produce real-time data from which 3D spatial features can be accurately modelled significantly in advance of mining. This innovation greatly expands on the information obtained from current in-seam directional drilling practices and provides for improved characterization of coal seams, including soft and weak zones, locations of geologic discontinuities, and analysis of formation flow.

This paper provides a description of the geosensing system and the interpretation of actual data from long in-seam directionally drilled methane drainage boreholes at mines in Australia, and discusses how this system increases the value of directional drilling as an exploration tool for the coal mining sector, and the evaporite and hard-rock mining sectors where directional drilling is also applied.

INTRODUCTION

Background

Underground in-seam directional drilling is performed extensively in Australian coal mines as the primary tool for methane drainage in gassy mines. Although effective for methane drainage, in-seam directional drilling also creates an opportunity to record valuable geological data, which has not been effectively realized due to limitations in existing underground in-seam directional drilling technology.

Historically, underground in-seam directional drilling has been performed with methane drainage in mind, and not subsurface exploration. Geological uncertainty remains the bane of many high-capacity underground coal operations. There are many examples in which large geological structures (such as faults, intrusions, etc.) or smaller outburst-prone features have been missed by in-seam drilling programs, only to be subsequently intercepted by continuous miners or longwalls. This has serious safety and productivity implications and severely limits the development of mining automation technologies without greater certainty in geological interpretation in advance of the face.

Currently, these issues exist at many operations due to ambiguous interpretations of surface-based geophysical data; and the lack of certainty in data generated by surface exploration drilling and underground in-seam directional drilling programs, the latter of which are entirely reliant on the driller's logs of "hard, soft, grey returns, coal/shale mix" or similar. Standards of reporting vary, and misinterpretation is common.

New developments in geosensing technology involve placing a series of real-time sensors onto existing underground directional drilling rigs, which integrate this data flow into the drilling process via interpretative software.