Assessment of Gold Mine Tailings as Based Geopolymer Binder in Concrete

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Abstract: With the increasing awareness of using industrial wastes in construction came the birth of the idea of geopolymerization. Geopolymerization is a process that produces a cementitious material with properties substantially comparable to those of conventional cement. Mine tailings (MT) have high silica content, which makes them eligible for geopolymerization. The use of mine tailing-based geopolymer binder as substitute for cement in concrete production will help address the disposal problem of these mine wastes. This preliminary study is aimed to investigate the viability of gold MT as a base of geopolymer binder for concrete. Assessment of its physical properties, chemical composition, microscopic structure, and unconfined compressive strength were performed to investigate its viability. The gold mine tailings-based geopolymer binder (MTGB) was produced using a combination of water glass solution (WGS) to 10 molar of sodium hydroxide (NaOH) solution ratio and alkaline solution (AS) to MT ratio. The mix proportion that produced the geopolymer binder cubes with the highest compressive strength were the AS-to-MT ratio of 0.35 and the WGS-to-10M NaOH ratio of 2.5.

Key Words: mine tailings; geopolymerization; mine tailing-based geopolymer binder

1. INTRODUCTION

The increasing worldwide demand for valuable materials such as metals, minerals, and coal resulted to enormous waste by-products that can cause environmental complications if not addressed properly. For example, valuable minerals from ores are obtained by crushing the parent rocks and leaving fine particles, known as mine tailings. These waste products which contain pollutants may undergo physical and chemical changes that are harmful to the atmosphere and nearby rivers and groundwater resources. This may further result to the contamination of food chains and potable waters that would eventually harm both animal and human health (Kossoff et al., 2014).

The improper disposal of these mine tailings in the country requires an immediate solution since failure to do so puts the lives of many people, especially those who live near mining sites, at risk. This is because mine tailings usually end up in large piles that are very unstable, prone to occurrences of landslides, and hazardous to the health of the communities nearby. Improper disposal of mine tailings has already caused disastrous incidents in the country throughout history. An example of the proper disposal technique may be the recycling of these wastes into useful construction materials, such as concrete. This idea is also supported by the studies of Suhendro (2014) and Adajar et al. (2017), which suggests the awareness of producing an environment-friendly concrete by incorporating industrial wastes in its production to be a fit substitute for conventional concrete.

The production of cement for concrete use alone has been proven by several studies to have detrimental effects on the environment. A study by Suhendro (2014) mentioned that 8% to 10% of the world's total carbon dioxide (CO2) emissions come from cement manufacturing. The carbon dioxide is released into the atmosphere when limestone and clays are crushed and heated to high temperatures.

With the increasing awareness on the idea of using industrial wastes in construction came the birth of the idea of geopolymerization. Geopolymerization is a process that produces a cementitious material with properties substantially comparable to those of a conventional cement; it occurs after an aluminosilicate material reacts with an alkali hydroxide or silicate solution (Ren, et al., 2014). Davidovits (2015) claimed that using geopolymer cement over OPC has less detrimental effects to the environment since the manufacture of this environment-friendly geopolymer cements through geopolymerization "does not require extreme high-temperature kilns, with large expenditures of fuel, nor does it require such a large capital investment in plant and equipment," and if applied in construction, it would likely reduce the carbon dioxide (CO2) emission of the construction industry by 80%.

According to Komnitsas and Zaharaki (as cited in Rao & Liu, 2015), geopolymerization is an integrated process for the synthesis of geopolymers, which involves leaching, diffusion, reorientation, polymerization, and condensation. Geopolymerization is triggered by the chemical reaction between an aluminosilicate material and an alkaline solution. The alkaline solution causes the dissolution of the aluminosilicate precursors and allows the molecules of the material to form three-dimensional networks with one another, then the mixture becomes an amorphous gel before it condenses and hardens into geopolymers or materials with cementitious properties similar to cement (Rao & Liu, 2015; Ahmari & Zhang, 2012). Most literature involving the production of geopolymer concrete use fly ash as the base material, however, the aluminosilicate material that was used in this study is gold mine tailings since some research showed that gold mine tailings have a high silica content, which makes it eligible for geopolymerization. Thus, this study is aimed to investigate the viability of gold mine tailings as a base for geopolymer binder intended to be used in concrete production.

2. METHODOLOGY

2.1 Gold Mine Tailings

The gold mine tailings (MT) sample was obtained from Itogon, Benguet, and was maintained in a dry condition before the experiment. The texture of the sample was soft and fine, with physical properties similar to cement powder. Preliminary tests using x-ray diffraction (XRD), x-ray fluorescence (XRF), and scanning electron microscope (SEM) analyses were performed on raw gold MT to determine the chemical compound and elemental composition, and average particle shape and size, respectively. The index properties were also identified such as specific gravity, grain size distribution, Atterberg limits, and linear shrinkage using ASTM D854, ASTM D422, ASTM D4318, BS 1377-2:1990 Part 6.5, respectively.

2.2 Aluminosilicate Material and Alkaline Solution

The geopolymer binder is produced through the combination of the aluminosilicate material and the alkaline solution. This is analogous to the production of cement paste, or the product of the combination of water and cement powder. The water glass solution (WGS) was composed of 14.65% of sodium oxide (Na_2O), 34.13% of silicon dioxide (SiO2), and 51.22% of water, while the 97%-98% pure sodium hydroxide pellets were dissolved in distilled water to produce the 10M of sodium hydroxide (NaOH) solution. The WGS-to-10M NaOH ratio used in the study of Aleem and Arumairaj (2012) was 2.50, while Patankar, et al. (2014a) used 1.00. Hence, the WGS-to-10M NaOH ratios that were used in this study were 1.00 and 2.50 because these values show to be the most frequently used in creating geopolymer concrete specimens.

The alkaline reagent produced by combining the 10M NaOH and WGS is recommended to be prepared at least 24 hours before use (Rangan, 2009; Davidovits as cited in Hardjito & Rangan, 2005). The ratio of the alkaline solution-to-aluminosilicate material affects the strength of the geopolymer concrete in such a way that, according to Patankar (2014), as the ratio increases, the strength also increases. The AS-to-MT ratio used in this study was 0.35 and 0.45 since they have been proven (Aleem and Arumairaj (2012), Patankar, et al. (2014a) and (2014b)) to produce geopolymer concrete specimens with adequate or considerable strength. The alkaline solution was mixed a day before the mixture of the wet and dry ingredients as suggested by Rangan (2009).

2.3 Geopolymer Binder

A preliminary procedure was done to determine the reaction of gold MT with the alkaline reagent to form a geopolymer and hence if it would harden. Some studies have shown to use copper mine tailings to successfully produce geopolymer bricks, however, it cannot be guaranteed that the same reaction would happen to the gold MT used in this study considering it was obtained from a different source and could be of a different composition, especially the level of aluminum and silica content, both of which are vital to the geopolymerization of the material.

The mine tailings-based geopolymer binder (MTGB) was produced through the combination of the aluminosilicate material and the alkaline solution. The MTGB sample with dimensions of 50x50x50mm cubes were produced, one for each combination of WGS-to-10M NaOH and AS-to-MT ratios and another using pure WGS as the reactor. The geopolymer binder specimens were hand mixed to ensure a more uniform consistency of the mixture. Four trial mixtures were produced based on the combination of the given ratios. An estimated total volume of 1.25 x10-3 m^3 of geopolymer binder per trial was needed as shown in Table 1 to produce a minimum of ten 50-mm mortar cubes for each trial mix.

Table 1. Mix Proportions for Geopolymer Binder

Materials	**Weight Proportions (kg)**			
Trial	**1a**	**1b**	**2a**	**2b**
WGS-to-10M NaOH	1		2.5	
AS / MT	0.35	0.45	0.35	0.45
Example Mix Design				
WGS	400	400	400	400
10M NaOH	400	400	160	160
MT	2285.71	1777.78	1600	1244.44

These specimens were heat cured for a temperature of 60^{o}C for 24 hours after casting in cube molds for at least 5 days since the specimens were still moist after few days of production. The total number of created specimens for geopolymer binders was 42, wherein at least 10 cube samples were made for each ratio. Five samples for each ratio were first tested on the 7th day, while the remaining samples were tested on the 28th day to determine the compressive strength of the MTGB. They were tested using the Unconfined Compression Test (UCT) machine which projects the applied load versus deformation curve. The mix ratio that produced the binder with the greatest compressive strength will be adopted for the production of the mine tailing-based geopolymer concrete.

3. RESULTS AND DISCUSSION

3.1 X-Ray Diffraction (XRD) and X-Ray Fluorescence (XRF) Analyses

The X-Ray diffraction analysis identified the presence of crystalline compounds in the mine tailings sample. However, the results of this analysis are purely qualitative and not quantitative. Only the presence of the compounds was determined, and not the specific amount or proportion contained in the sample. Peaks on the XRD graph, shown in Figure 1, suggest that the material is crystalline, while if the lines are somehow flat, the material is amorphous. The gold MT used in this study contain crystalline compounds such as illite, kaolinite, montmorillonite, and quartz. The presence of Quartz has a chemical composition of SiO2 which shows the presence of silica compounds in the sample.

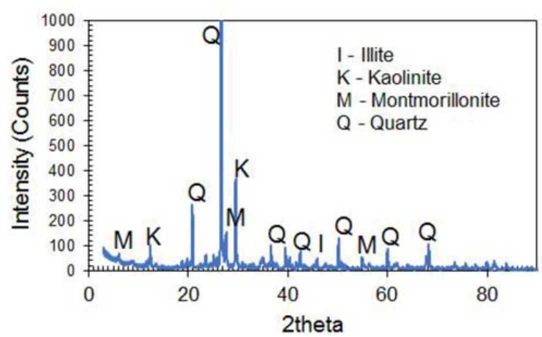

Fig. 1. XRD Results of Mine Tailings

The major elemental components of MT were identified using the XRF analysis, the result is shown in Table 2. The top two major elemental components are silicon (Si) and aluminum (Al). Comparing the result to other related literature, the MT used in this study have more aluminum but less silicon content. The Si-to-Al ratio of the MT is 4.81. The silicon (Si) and aluminum (Al) content of the MT is important in the geopolymerization process. The presence of aluminum and silicon compounds in gold MT makes the gold mine tailings from Itogon, Benguet viable for geopolymerization.

Table 2 Elemental Composition of Gold MT

Elements		(%)
Al_2O_3	**Aluminum oxide**	**12.867**
As_2O_3	Arsenic trioxide	0.064
CaO	Calcium oxide	5.731
Cr_2O_3	Chromium(III) oxide	0.008
CuO	Copper(II) oxide	0.009
Fe_2O_3	Iron(III) oxide	5.762
Ir_2O_3	Iridium sesquioxide	0.003
K_2O	Potassium oxide	4.546
MgO	Magnesium oxide	1.752
MnO	Manganese(II) oxide	0.357
PbO	Lead(II) oxide	0.011
RbO	Rubidium oxide	0.017
SO_3	Sulfur trioxide	6.335
SiO_2	**Silicon dioxide**	**61.848**
SrO	Strontium oxide	0.015
TiO_2	Titanium dioxide	0.587
V_2O_5	Vanadium(V) oxide	0.033
ZnO	Zinc oxide	0.047
ZrO_2	Zirconium(IV) oxide	0.011

3.2 Scanning Electron Microscopic Analysis

Scanning Electron Microscopic (SEM) analysis was performed to determine the microstructure of he gold MT. Based on the results of the SEM analysis, shown in Figure 2, it can be seen that most of the particles of the mine tailings sample are flaky. Flaky particles are usually associated to the presence of clay minerals.

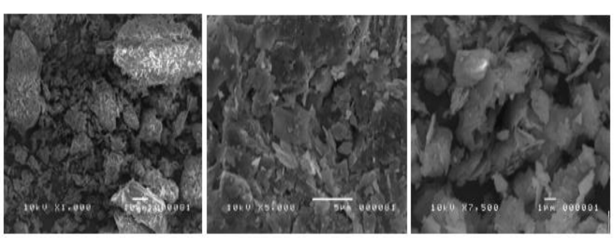

Figure 2. 1000X, 5000X, and 7500X magnification from SEM test

Flaky particles affect the durability of concrete as they cause laminations tending to orient in one plane only (Manjunath and Chandra, 2014). Furthermore, as cited in Manjunath and Chandra (2014), Chen et al. mentioned those flaky particles are more susceptible to breakage, while Sakthibalan discussed those flaky particles produce more voids and thus, result in low workability and strength.

3.3 Index Properties of Mine Tailings

The specific gravity of the gold MT was determined using ASTM D854 and has a value of 2.63. The obtained specific gravity is within the range of values for mine tailings obtained from the study of Adajar (2014). The Atterberg Limits of the gold MT sample were based on ASTM D4318. The linear shrinkage of the sample was based on BS 1377-2:1990 Part 6.5. The values of Liquid Limit (LL), Plastic Limit (PL), and Plasticity Index (PI) are shown in Table 3.

Table 3. Index Test Results of Mine Tailings

Index Test	
Specific Gravity, Gs	2.63
Liquid Limit, LL	27
Plastic Limit, PL	22
Plasticity Index, PI	3

The grain size distribution curve (GSDC) of the gold MT is shown in Figure 3. The very steep GSDC indicates that the sample is uniformly graded. Based on Unified Soil Classification System (USCS), the gold MT sample may be classified as inorganic silt and very fine sand with low plasticity (ML). The average particle size (D_{50}) of the gold MT is 0.15mm. The sample has coefficient of uniformity (Cu) value of 2.47 and coefficient of curvature (Cc) value of 1.93.

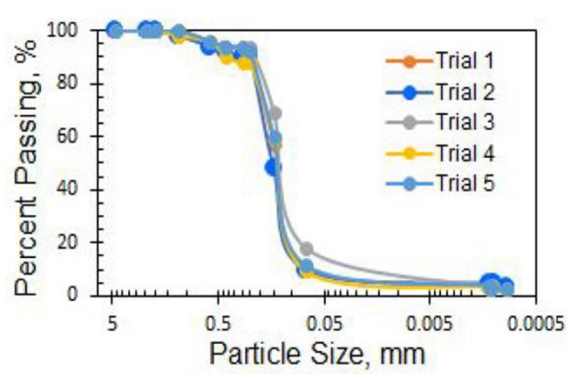

Figure 3. GSDC of Gold MT

3.4 Mine Tailings-Based Geopolymer Binder (MTGB) Compressive Strength

From the performed unconfined compression test (UCT), results shown in Table 4, it was observed that Binder 2a possessed the highest unconfined compression strength both on the 7th day (Figure 4) and 28th day (Figure 5), with a value of 1.541 MPa, and 1.636 MPa, respectively.

Table 4. Unconfined Compression Strength of Mine Tailings Geopolymer Binder

Sample ID	Unconfined Compression Strength, MPa	
	Day 7	Day 28
Binder 1a	1.225	1.626
Binder 1b	0.831	1.362
Binder 2a	1.541	1.636
Binder 2b	1.155	1.237

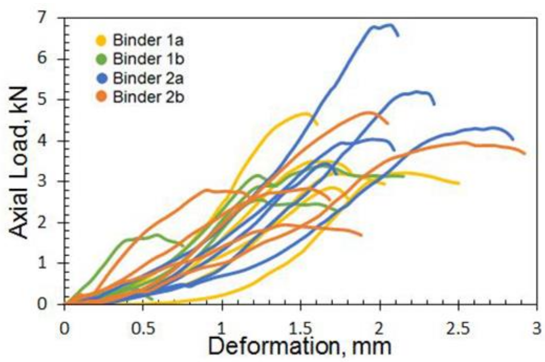

Figure 4. 7th Day Compressive Strength of MTGB

Binder 2a has an AS-to-MT ratio of 0.35, and a WGS-to-10M NaOH ratio of 2.5. Comparing the two ratios for AS-to-MT of 0.35 and 0.45 shows that the amount of alkaline solution, which is 35% of the number of mine tailings, is enough to produce a binder. However, increasing the amount of solution to 45% decreases the strength of the binder. WGS, which is used as a binder or adhesive, is required to have a greater amount than the 10M NaOH. This is to ensure the adhesivity of the binder and its ability to set. On the other hand, 10M NaOH is also used to increase the strength of the binder, but excessive 10M NaOH may result in the occurrence of efflorescence and this was observed in the physical appearance of the sample. This is the result of some of the sodium hydroxide solution not reacting completely with the mine tailings. Geopolymer binder samples with efflorescence have lower strength values compared to the samples without efflorescence (Zhang, et al., 2014). Moreover, the specimens produced with a higher amount of water will result to a higher porosity. This may lead to the formation of higher efflorescence products within the pores, which then translates to higher inner stress. This eventuality could account for the low strength of the geopolymers with efflorescence.

For the 28th-day unconfined compression test, as shown in Figure 5, the load-deformation curve showed a steeper slope indicating the increased stiffness and higher strength of the samples in comparison to that of the 7th day strength.

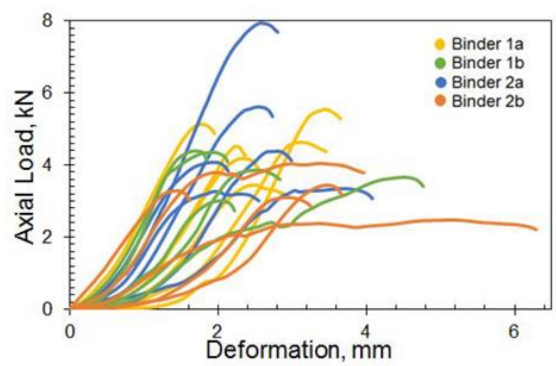

Figure 5. 28th Day Compressive Strength of Mine Tailings Geopolymer Binder

Binders 1b and 2b show a lesser value of maximum applied axial load thus lesser unconfined compressive strengths. These two mixes, Binders 1b and 2b, with the AS-to-MT ratio of 0.45 showed that too much alkaline solution may result in a more workable mixture, but binder specimens with lesser compressive strength. The amount of alkaline solution must be adequate to produce the geopolymerization. Binders 1a and 2a presented lesser steep graphs, but binder 2a resisted a greater load. Thus, the ideal ratio combination that is expected to produce higher compressive when used as the mine tailings-based geopolymer concrete is Binder 2a, which consists of 0.35 as AS-to-MT ratio and 2.5 as WGS-to-10M NaOH.

4. CONCLUSIONS

Preliminary assessment of gold MT as based-geopolymer binder was undertaken to determine its viability in the production of geopolymer concrete. The index properties of gold MT from Itogon, Benguet revealed that the mine tailing is fine-grained soil having particles with roughly the same sizes or shapes and may then be considered as uniformly graded inorganic silt with low plasticity (ML) with a specific gravity of 2.63. The average liquid limit of the gold MT was found to be 27, while the average plasticity index was 3; on another hand, the plastic limit and linear shrinkage were found to be 22 and 3, respectively.

The gold MT used in this study contain crystalline compounds such as illite, kaolinite, montmorillonite, and quartz. quartz and muscovite which both have chemical constitutions containing aluminum and silicon compounds. The two major elemental components found in the gold MT are silicon (Si) and aluminum (Al), with Si-to-Al ratio of 4.81 The presence of aluminum and silicon compounds in gold MT makes the gold mine tailings from Itogon, Benguet viable for geopolymerization. The microstructure of gold MT showed flaky particles associated to the presence of clay minerals.

The MTGB was produced using four proportion combinations of WGS-to-10M NaOH and AS-to-MT ratio. The unconfined compression strength of MTGB was obtained to determine which mix proportion produced the highest compressive strength. The highest compressive strength obtained among the tested binders had a WGS/NaOH ratio of 2.5 and an AS/MT ratio of 0.35, with a value of 1.541 MPa on the 7th day, and 1.636 MPa on the 28th day.

Thus, it can be concluded that the MTGB with WGS/NaOH ratio of 2.5 and an AS/MT ratio of 0.35 is viable as binder to substitute cement in the production of geopolymer concrete.

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