Obtaining A Cementitious Geopolymer From Gold Mining Tailings With Possible Use In Engineering Applications

Walter Pardavé[1]*, Manuel Epalza[1], Jorge Durán[2], Daniel F. Lóvera[3]

1 Environmental Group of Applied Research, Universidad de Santander, UDES, Bucaramanga, Colombia.
2 MetallurgicalEngineer, Universidad Industrial de Santander, UIS, Bucaramanga, Colombia.
3 Department of Metallurgical Engineering, Universidad Nacional Mayor de San Marcos, Lima, Perú
Corresponding author: Walter Pardavé

ABSTRACT:*The aim of the present study was to obtain a substitute of Portland cement that implies environmental benefits. According to the above, a cementitious geopolymer was synthesized, at laboratory scale, from the alkaline activation of gold mining tailings from the department of Santander, Colombia. This material was exposed to a series of pre-treatments which included the removal of impurities and moisture; after sieving the material corresponding to the 100-mesh Tyler standard sieve was selected, which was characterized by means of X-ray diffraction (XRD) and X-ray fluorescence (XRF) techniques. In addition to the mining tailings, high purity kaolin and alumina were used as precursor materials. These were exposed to a pre-heat treatment at 600°C for one hour. The activating solution consisted of a mixture of sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3) in a weight ratio of 7:3. After mixing the precursor material with the activator solution, by mechanical stirring, the initial curing was performed at 90°C for 24 hours. The performance of the obtained product was determined by the evaluation of its compressive strength under standard conditions and at high temperatures according to the curing time. In addition, a scanning electron microscope (SEM) coupled with a dispersive energy spectrometer (EDS) was used to identify microstructural characteristics of the geopolymer. This presented properties like Portland cement I, so the use of this type of products in engineering applications could be a good option for the treatment of mining tailings and, in turn, the reduction of the pollution produced by the manufacture of the Portland cement.*

KEYWORDS *Cementitius, geopolymer,gold, mining, tailings*

I.   INTRODUCTION

The availability of raw materials and its ease of production and application have made Portland cement the most popular and widely used construction material worldwide [1, 2]; with an annual consumption of more than 1000 Mt of Portland cement, water is the only one used in greater quantity by mankind [2]. Portland cement has been used as a binding agent for concrete due to its exceptional thermal performance, mechanical properties and durability [1]. In addition, the application of concrete in infrastructure and transport has undoubtedly improved the quality of life of civilization and boosted its development [2]. However, there are environmental difficulties associated with its use: a huge energy consumption (3630 MJ/t) and the emissions of carbon dioxide (CO_2) during its manufacture [3, 4, 5]. The production of one ton of Portland cement requires 2.8 tons of raw materials and fuels, and approximately 0.8 tons of CO_2 are created because of the combustion of fossil fuels and the thermal decomposition of calcium carbonate to produce silicates of calcium and aluminates, the base of the Portland cement [4, 2, 6]. Overall, about 13.5 billion tonnes of CO_2 are produced from global cement production, accounting for approximately 8% of annual greenhouse gas emissions [7, 8].

On the other hand, materials called geopolymers have increased their popularity in the construction industry, in the last years, because they are ecologically friendly, have a low cost, allow the encapsulation of hazardous wastes and the energy required during their production is low (990 MJ/t) [9, 10, 5]; the use of these materials significantly reduces CO_2 emissions [11] up to 80% compared to Portland cement [6, 12]. A geopolymer is an inorganic cementing material that is produced by an alkaline activation process: the reaction

between a solid aluminosilicate (precursor) in a highly alkaline solution (activator) that produces an inorganic polymerization [5]. This sort of materials is well known for having high compressive strength, high temperature stability, low thermal conductivity, among other properties of interest in engineering [13, 14, 15].

As precursors of the geopolymerization reaction, different raw materials based on aluminosilicates have been used, including industrial solid wastes [16, 17]. However, fly ash, blast furnace slag and metakaolin are the main precursors used in geopolymer production [16].

Because mining wastes have negative environmental impacts resulting from their accumulation, including removal of vegetation cover, deforestation, changes in land slopes, increased risk of erosion, contamination of water and agricultural products and risks to human health [18] and, in addition, few studies have been carried out in the case of wastes from gold mining as precursors of geopolymers, the aim of this study was to obtain a geopolymer using gold-mine tailings from the department of Santander, Colombia, as a precursor. Considering the different chemical characteristics of the gold minerals [19], the main physicochemical characteristics of the geopolymer obtained will be evaluated to determine the feasibility of its use as cementitious material in engineering applications.

II.   METHODOLOGY

2.1 Materials
In addition to kaolin and high purity alumina, a sample of gold mining tailings from the department of Santander, Colombia, was used as precursor material. The mining tailings were washed with a slightly acidic solution and water to remove impurities and soluble salts that could affect the geopolymerization. Subsequently, this material was exposed to gravimetric and magnetic separation processes to extract iron oxides, compounds of inert nature in the geopolymerization process, to improve the performance of the alkaline activation. Finally, it was left outdoors for a week to remove excess moisture, which restricts the screening process; the material corresponding to the 100-mesh Tyler standard sieve was selected.

The activating solution consisted of sodium hydroxide (NaOH) and sodium silicate (Na2SiO3) in a weight ratio of 7:3. NaOH enables dissolution of aluminosilicate sources while Na2SiO3 acts as activating agent, binder and plasticizer for geopolymers [20].

2.2   Characterization of gold mining tailings
Once the treatment described above was done to mining tailings, this material was characterized by means of X-ray diffraction (XRD) and X-ray fluorescence (XRF). In the case of XRD, it was operated a Bruker D8 Advance diffractometer with an incident wavelength corresponding to the copper $K\alpha1$ radiation (1.5406 Å); sweeps were performed from $2\theta=3.5°$ to $2\theta=70°$ at 0.03 2θ·s-1 of speed, 40 kV of voltage and 30 mA of current. Phase identification was carried out by means of the International Center for Diffraction Data (ICDD) database. The XRF analysis was performed on a Bruker S8 Tiger equipment with a Rh target tube.

2.3   Synthesis of geopolymers
The precursor material was mixed in a mortar (mining tailings, kaolin and alumina in a weight ratio of 2:2:1) until a homogeneous mixture was obtained. This mixture was then placed in a clay pot and heated at 600°C in a muffle for one hour for subsequent cooling to room temperature (25°C).
The precursor material and the activating solution were mixed on a mechanical stirrer for 8 min. The mixture was placed in square acrylic molds of 50 cm of side, previously greased and considering the specified in the ASTM C109/C109M 16a standard, for its later curing at 90°C for 24 hours. After three days, the specimens were wrapped in polyethylene films to isolate them from environmental conditions that could influence their properties, such as humidity.

2.4   Characterization of products
Compression tests were performed according to ASTM C109/C109M 16a at 7, 14 and 28 days of curing (see Figure 1). In addition to the compressive strength under standard conditions, the strength of the test specimens was also determined after being exposed to high temperatures (300-1000°C) for 2 hours; each determination was performed in triplicate. These tests were also carried out on specimens made with Portland cement type I to study the feasibility of replacing it with the obtained geopolymer.
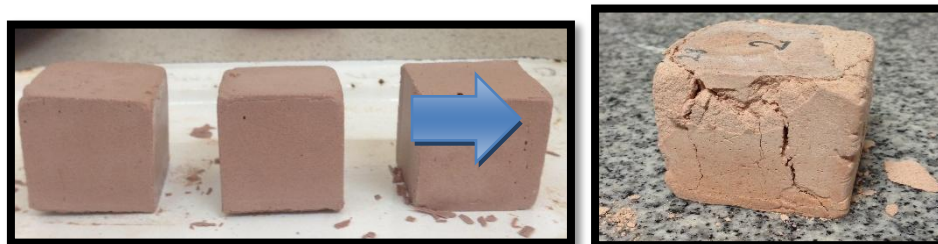
Figure 1. Results of Compression tests were performed according to ASTM C109/C109M 16a at 7, 14 and 28 days of curing

III. RESULTS AND DISCUSSION

The phases identified by XRD in the gold mining tailings are shown in Table 1. Likewise, the elemental chemical analysis and the phases associated with each element quantified, which was obtained by means of XRF, can be seen in the Table 2. In both cases, a majority fraction of SiO2 and Al2O3 is observed, complemented by other aluminosilicates such as muscovite and orthoclase, phases that play a fundamental role in geopolymerization[5]. In addition, the mineral also has a representative fraction of Fe2O3, usually present in some clays used to produce geopolymers, but in a slightly lower percentage [21].

Table 1. Phases identified by means of XRD in the mining tailings used to obtain a cementitious geopolymer.

Phase Name	Chemical formula	Concentration (wt. %)	JCPDS-ICDD card
Quartz	SiO_2	52.6 ± 0.2	010-87-2096
Corundum	Al_2O_3	20.2 ± 0.1	010-89-7717
Muscovite	*$KAl_2Si_3AlO_{10}(OH)_2$*	*15.1 ± 0.2*	*000-07-0032*
Orthoclase	$K(AlSi_3O_8)$	10.3 ± 0.1	010-75-1190
Hematite	Fe_2O_3	1.8 ± 0.1	010-73-2234
Microcline	$K(AlSi_3O_8)$	NQ*	010-70-6187

* NQ: Not Quantifiable.

Table 2. Quantitative chemical analysis of the mining tailings used to obtain a geopolymer by means of XRF.

Element (wt. %)[a]															
Si	Al	K	Fe	Mg	Ti	Na	Ba	Ca	P	Zr	Cu	S	Zn	As	Rb	V
27.73	17.51	2.53	1.45	0.38	0.31	0.11	0.07	0.03	0.02	0.02	0.02	0.02	0.01	0.01	96*	87*

Oxide (wt. %)[b]															
SiO_2	Al_2O_3	K_2O	Fe_2O_3	MgO	TiO_2	Na_2O	BaO	CaO	P_2O_5	ZrO_2	CuO	SO_3	ZnO	As_2O_3	Rb_2O	V_2O_5
59.32	33.09	3.05	2.07	0.63	0.51	0.15	0.08	0.04	0.05	0.03	0.02	0.05	0.02	0.02	0.01	0.02

[a] Balance: C, H, O. Sc was detected but could not be quantified. * ppm.

[b] Samples were calcined at 950°C for two hours to determine ignition losses (0.8%).

Concerning to the obtained geopolymer, Figure 2 shows its compressive strength according to the curing time; the strength of the Portland cement I is also shown by way of reference. Initially, the strength of the Portland cement is considerably higher than that of the geopolymer. However, as the cure time elapses, the strength of the latter is approximately equal, which is an indication that it could replace Portland cement in some applications.

In the case of compressive strength after exposure to elevated temperatures, the geopolymer again exhibits a behaviour like Portland cement, as can be observed in the Figure 3. However, highlights a slightly superior strength of the geopolymer at 800°C. Recrystallization at high temperatures to form heat-resistant phases such as nepheline and albite from hydroxysodalites which might be present in the geopolymer, clearly did not occur [22].
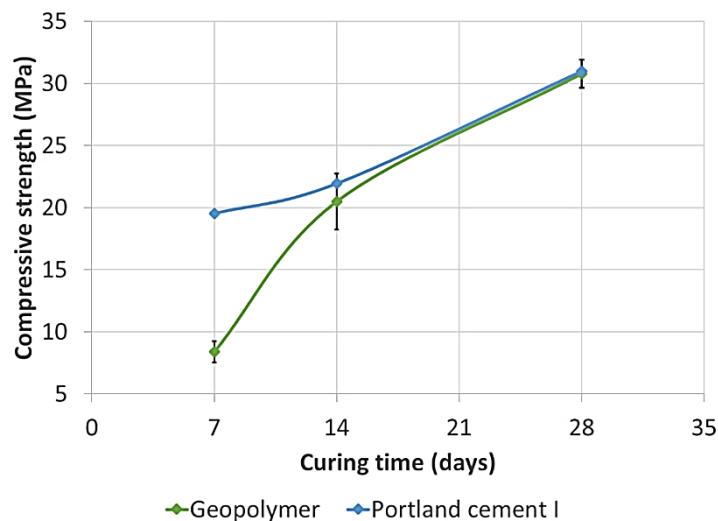
Figure 2. Comparative compressive strength of geopolymer and Portland cement I according to curing time.
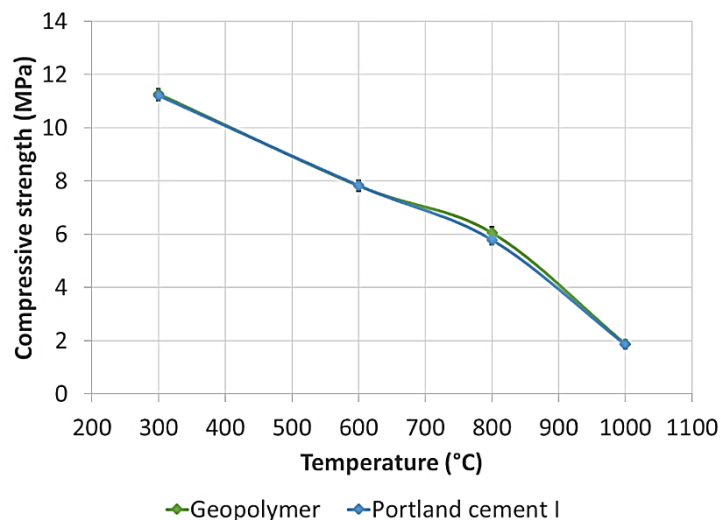

Figure 3. Comparative compressive strength of geopolymer and Portland cement I after being exposed to several high temperatures for two hours.

Although several researchers have found that geopolymers have more strength than Portland cement, this fact depends on several factors. For example, the increase of concentration in NaOH solution could improve the performance of geopolymers in compression tests because it implies a better dissolution of aluminosilicates and, in turn, a better mechanical behavior of the geopolymer [23, 24]. The initial curing extended at room temperature, instead of temperatures above 50°C, also contributes to improving the final strength of the geopolymer [25, 26]. Increasing the amount of soluble silicates (such as Na_2SiO_3) would also increase the mechanical strength of geopolymers due to increased gel production [27]. The low strength of the geopolymer could be attributed, in part, to the fact that the Si/Al ratio of the mineral has a value of 1.6, according to the results obtained by XRF; the optimal value of this ratio to obtain a superior compressive strength would be 1.9 according to some authors [28].

On the other hand, Figure 4 shows the results of the SEM-EDS analysis performed on a geopolymer sample after 3 days of curing time. Unlike other studies on the subject [24], the presence of a gel phase cannot be detected in the SEM image, which may be associated with the difficulty of overcoming the strength of Portland cement I [27].
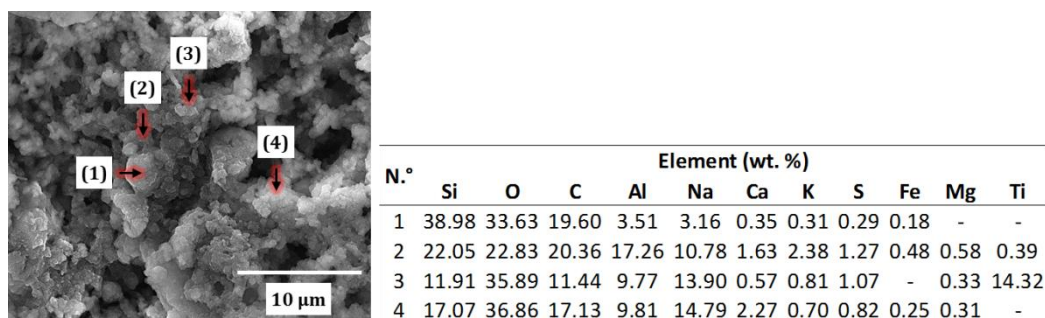
Figure 4. Geopolymer SEM-EDS analysis after 3 days of curing time. The table summarizes the weight percentage of each element corresponding to areas identified on the SEM secondary electron image. These were obtained through semi-quantitative chemical microanalysis by means of EDS.

IV.　CONCLUSIONS

A cementitious geopolymer with mechanical properties like Portland cement I was obtained. The replacement of the latter with this geopolymer in some engineering applications would not only reduce the environmental pollution generated by Portland cement production, but also that produced by the gold mining.

ACKNOWLEDGEMENTS

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