Stabilization of an iron ore tailings by-product with perlite waste geopolymer

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ABSTRACT

In the field of engineering, sustainable solutions that can generate lower environmental impacts, either through material replacement or reuse, are increasingly sought after. In Brazil, in the geotechnical area, there is a demand to find solutions to avoid the disposal of tailings sludge destined for structures, following the recent dam ruptures of Fundão in 2015 and Córrego do Feijão in 2019. One of the recent investments is in the treatment of tailings, generating by-products. To improve the mechanical properties of these by-products, geopolymer utilization has been employed. Geopolymers are products resulting from reactions between aluminosilicate precursors and alkaline activators. The objective of this study is to evaluate the unconfined compressive strength of an iron ore tailings by-product stabilized with a geopolymer that utilizes perlite waste as a precursor and sodium hydroxide as an activator through a two-part alkali-activation process. Two molar concentrations of activators, 2M and 5M, and two sample compositions were evaluated: one with 20% geopolymer and 80% iron ore tailings by-product, and another with 30% and 70%. An increase in unconfined compressive strength was observed with higher concentrations of the activator solution and a greater percentage of geopolymer in material stabilization. Thus, it can be stated that the use of geopolymers in by-product stabilization is promising, but finding the optimal dosage and evaluating other variables such as curing time and temperature is necessary.

Keywords: Soil stabilization; mining tailings; perlite waste; geopolymers.

1. Introduction

Since the recent ruptures of the Brazilian tailings dams of Fundão in 2015 and Córrego do Feijão in 2019, the eyes of researchers and specialists have turned to these structures, in order to evaluate the construction methods of active dams and propose methodologies for new constructions. Regarding the construction methods, Law 14.066 of 2020 (BRASIL, 2020) was sanctioned, requiring that all dams built by the upstream method be decommissioned and also prohibiting new dams to be built by this method, since it was considered to have a lower safety factor due to the lack of control of the water table near the downstream slope (Soares, 2010). There are some methods for the deactivation of dams that have already been built upstream, such as decharacterization, which is the total or partial removal of the tailings from the dam, which then undergoes a process of moisture removal so that they can be stacked dry, or the waterproofing of the entire lagoon. Many times, both methods require the concreting of this material so that the structure is not compromised by the entry of water.

The introduction of cement in the mass of material provides improvements in the mechanical properties of the mining waste, however, cement in its production process spends a large amount of energy and emits large amounts of CO_2 into the atmosphere. According to the Global Cement and Concrete Association (GCCA, 2020), 5 to 8% of global CO_2 emissions come from cement plants, and according to Benhelal et al. (2013), who already pointed out a share of 5 to 7% of global CO_2 emissions in cement manufacturing, they expressed that to produce 1 ton of cement 900 kg of CO_2 are emitted into the atmosphere.

An alternative to the use of this material is the manufacture of geopolymers. Studies involving alkaline activation reactions began in the 1930s, but it was only in the 1970s that the French scientist Joseph Davidovits gave the nomenclature geopolymers, commonly used until today. Geopolymers are materials that result from chemical reactions between a precursor, rich in silica and alumina (such as ashes, metakaolin, blast furnace slag and residues from other materials) and an alkaline activator, the most common being sodium hydroxides and silicates. In the end, a new material is obtained, with greater resistance to acids and sulfates (Aiken et al., 2017; Albitar et al., 2017; Bakharev, 2005), heat (Kong & Sanjayan, 2010; Sarker et al., 2014) lower drying and wetting shrinkage (Sagoe-Crentsil et al., 2013) and mechanical strength (Livi, 2013; Luukkonen et al., 2018; Chanta & Palazzi, 2017; Figueiredo et al., 2021; Segura et al., 2022). In this scenario, aiming at environmental and human health impacts, alternative materials to cement are sought in soil stabilization and geopolymer manufacturing.

The production of perlite, a material widely used as a filtration element for substances in pharmaceuticals and in plants, generates waste - about 20% of the total production (Burgos, 2019) - with potential to meet these two demands, being minerals of volcanic origin rich in silica and alumina, indispensable composition in the precursor materials of geopolymers. Chanta & Palazzi (2017), Burgos (2019) and Ribeiro (2019) have already evaluated some properties of the material in soil stabilization, in the manufacture of bricks and blocks in social housing and in the removal of oils from vehicle washing, respectively, and obtained significant results regarding the strength of the materials and the removal of the contaminant.

Therefore, the objective of this study was to evaluate the unconfined compressive strength of samples of mining tailings by-products stabilized with perlite waste-based geopolymers. Four samples with different molar concentrations of the activator solution and geopolymer contents in the mixture were evaluated.

2. Geopolymers

Geopolymers are formed by means of alkaline activation reactions, in which one or more precursors (usually ashes, metakaolin, blast furnace slag or waste with high contents of silica and alumina) come into contact with activators (most commonly used sodium silicates and hydroxides).

According to Palomo (1999) these are chemical processes that allow the transformation of certain partially or totally amorphous structures into others with cementitious properties. In the first stage of alkaline activation there is an exothermic dissolution that promotes the breaking of the covalent bonds between the oxygen, aluminum and silicon atoms, originating the silica and alumina ions; In the second stage, there is what is called the equilibrium phase, in which the product resulting from the previous phase accumulates for a given time without releasing heat, after that, the molecules go through the stages of condensation and reorganization in which they begin to form a gel, and in the final stage, geopolymerization, there is the formation of three-dimensional structures, whose formed gel acts as a binder, giving greater mechanical strength to the particles (Castro, 2015 - see Fig. 1). In the literature, besides the gain in mechanical strength pointed out by several authors (Van Jaarsveld et al., 1997; Davidovits, 1988; Atis et al., 2015), there is an increase in resistance to acid and sulfate attacks (Van Jaarsveld et al., 1997; Davidovits, 1988; Vance et al, 2009), to freeze-thaw cycles (Van Jaarsveld et al., 1997), low hydraulic conductivity (Castro, 2015), structural stability when subjected to high temperatures (Davidovits, 1988; Vance et al., 2009; Castro, 2015) among others.

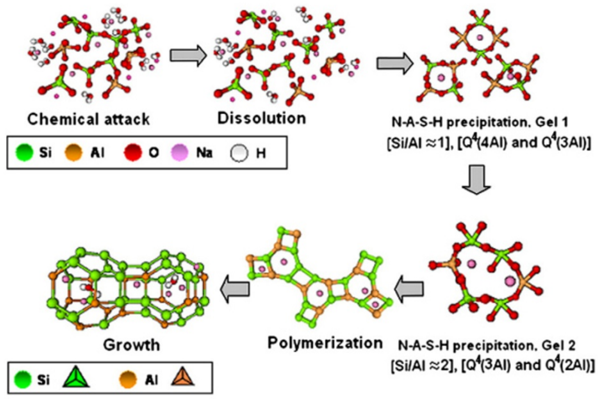

Figure 1. Alkaline activation steps. Source: Shi et al. (2011).

There are two ways of manufacturing geopolymers (see Fig. 2), which differ in the addition of water to the mixture. Two-part geopolymers are the most common, already in large-scale use, in which solid precursors are mixed with activators in solution, then usually more water is added to ensure the workability of the geopolymer paste. One-part geopolymers, on the other hand, are made by mixing the solid precursors and activators, and only then water is added. So, while in one-part geopolymers the water is added only once, in two-parts it is added in two steps. In this study, geopolymers were manufactured by the two-parts method, in which an activator solution was used.

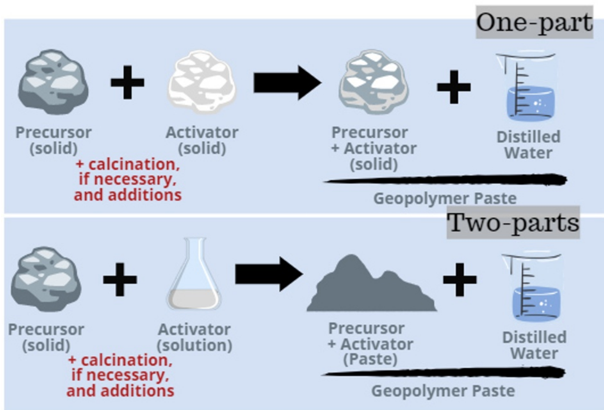

Figure 2. Geopolymer manufacturing methods.

There are several variables that can influence the mechanical strength of geopolymers, such as the Si/Al and Na/Al ratios, liquid/solid ratio, activator concentrations, curing time, temperature, among others. About the Si/Al and Na/Al ratios, according to Khale & Chaudhary (2007), the high content of reactive silica involves the formation of a large amount of alkaline aluminum silicate gel and, consequently, a high mechanical resistance is developed. Davidovits & Sawyer (1985) already pointed out, in their patent number 4.509. 955, the appropriate ratios of SiO_2/Al_2O_3 between 3.3 to 4.5 and M_2O/Al_2O_3 from 0.8 to 1.6M being a metallic cation, however, according to Khale & Chaudhary (2007), when waste is used as precursor, this range may vary, what is indicated is that the M_2O/Al_2O_3 ratio be close to 1, because very high alkaline concentrations may cause efflorescence.

3. Materials

3.1. Perlite waste

The perlite residue (see Fig. 3) used in the research was obtained from the company Buntech, is granular and went through milling until it reached the particle size of material passing the sieve #200, of 0.075 mm in order to react better with the chosen activator. The actual density of the grains is 2.16 g/cm³.

Figure 3. Perlite waste.

In the mineralogy of the material (see Fig. 4), it is possible to perceive a large amount of Quartz (represented by Q in the Fig. 4), Anorthite (A) and, to a lesser extent, Biotite (B).

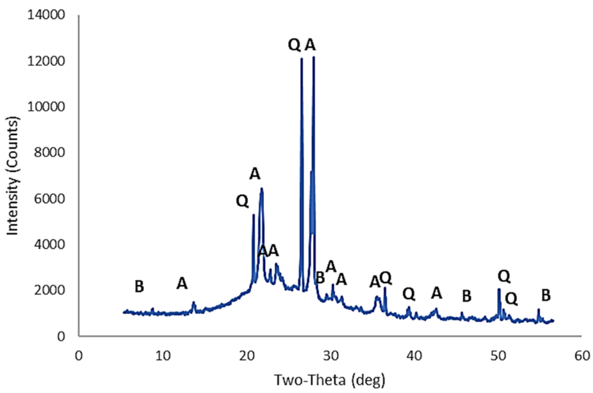

Figure 4. Mineralogy of perlite waste.

3.2. Alkaline activator

The activator chosen for alkaline activation was Sodium Hydroxide (NaOH), aiming the Na/Al ratio and the cost of geopolymers. The solid sodium hydroxide, in micro beads, was diluted in distilled water to obtain different concentrations: 2 and 5M.

3.3. Iron ore tailings by-product

The material stabilized with the geopolymers was an iron ore-based by-product obtained by Vale (see Fig.5). The company developed a technique to produce sand using the same iron ore production process, only adding new stages of concentration, classification and filtration, thus reducing the volume of tailings discarded in piles and dams (Vale S.A., 2021). The actual density of the by-product grains is 2.69 g/cm³.

Figure 5. Iron ore tailings by-product.

In the mineralogy of the material (see Fig. 6), it is possible to perceive a large amount of Quartz (Q) and Hematite (H) and, to a lesser extent, Charmosite (C) and Biotite (B).

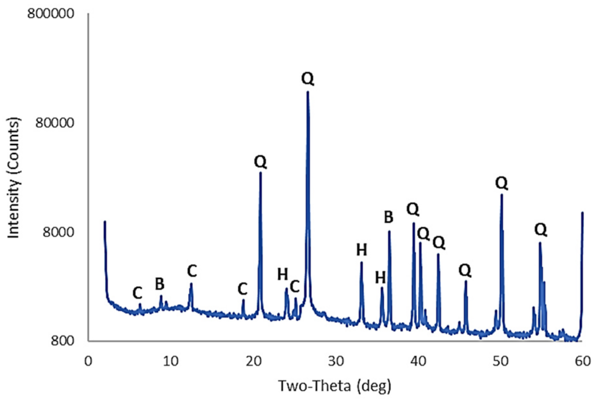

Figure 6. Mineralogy of by-product.

In Table 1 it can be seen the results of the chemical analysis of the materials.

Table 1. Results of the chemical analysis.

Sample	SiO_2 (%)	Al_2O_3 (%)	K_2O (%)	Na_2O (%)	Fe_2O_3 (%)
Perlite Waste	76,59	12,11	2,26	3,98	2,88
By-product	90,38	0,42	0,21	0,02	14,12

4. Methodology

Initially, chemical and x-ray diffraction analyses were performed to know the chemical compositions and mineralogies of the materials involved. Then the specimens were molded, compacted, and tested by means of the unconfined or simple compression strength tests.

To determine the Unconfined Compressive Strength (UCS), the NBR 12770 standard (ABNT, 1992) was used, which consists in applying an axial load on the samples. The specimens used have 5 cm in diameter and 10 cm in height and were obtained by means of compaction with normal energy. The test speed was 1.27 mm/min. For a better comparison between the composites and to obtain the complete curve, the test was performed up to 3 minutes, even though rupture had already occurred. It was sought to continue the test to evaluate the post-peak and thus understand the behavior of the samples after rupture. The unconfined compressive strength was calculated according to Eq. (1):

$$UCS = \frac{P}{\frac{100*Ai}{100-\varepsilon}} \quad (1)$$

Where: P is the applied load; Ai is the average cross-sectional area; ε is the specific axial strain corresponding to the loading.

It was not possible to perform the test only with the by-product because it is a non-plastic, cohesionless material.

In this study the following variables were evaluated:

- Molar concentration (2 and 5 mol/L);
- Composition of the samples (amount of geopolymer in the stabilization of the iron ore tailings by-product - 20 and 30%).

In all samples a liquid/solid ratio of 0.8 was used and the by-product was in a dry state, the curing time was 7 days, and, in the case of the precursors, perlite waste and the iron ore tailings by-product itself were added as an additional source of silica and alumina.

In the case of the Si/Al and Na/Al ratios, as molding moistures were not evaluated for the stabilized material, water was added only once in the mixture: in the activator solution. In the cases of the first two samples, whose concentrations are 2M, the values of the proportions were 5.1 and 0.6, respectively, whereas in the other two samples, with a concentration of 5M, the values were 5.4. and 0.7.

5. Results

Fig. 7 shows the results obtained from the unconfined compressive strength tests of four samples of the iron ore by-product stabilized with geopolymers.

It is possible to observe an increase in strength with increasing molar concentration of the activator solution, when comparing samples 2M_20%GP+80%by-product (34,11 kPa) and 5M_20%GP+80%by-product (254,33 kPa) and samples 2M_30%GP+70%by-product (59,16 kPa) and 5M_30%GP+70%by-product (1129,37 kPa). Comparing samples 2M_20%GP+80%by-product (34,11 kPa) and 2M_30%GP+70%by-product (59,16 kPa) and samples 5M_20%GP+80%by-product (254,33 kPa) and 5M_30%GP+70%by-product (1129,37 kPa) also shows an increase in strength with increasing percentage of geopolymer inserted into the mixture. The difference in results among the samples can be observed more clearly in Fig. 8, which shows the peak resistances in relation to the geopolymer content.

In terms of material behavior, it is found that the increase in molar concentration and geopolymer content causes the samples to show greater brittleness, presenting a higher peak strength, but which does not hold with greater deformations.

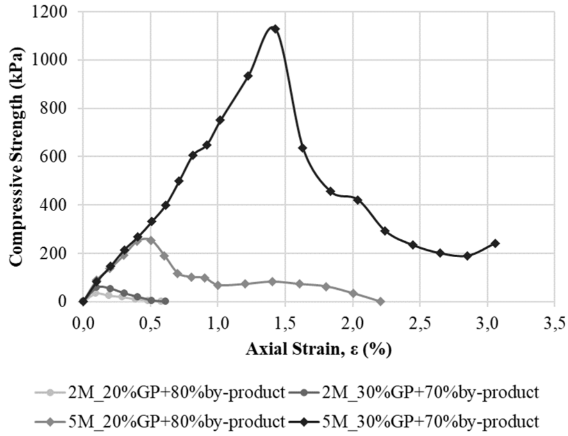

Figure 7. Results from the Unconfined Compressive Strength tests.

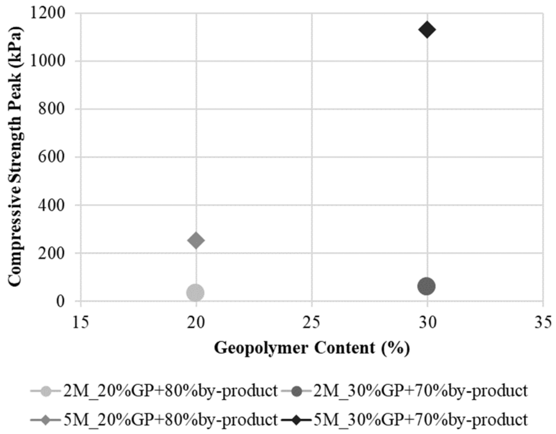

Figure 8. Results from the Unconfined Compressive Strength peak.

One of the few studies that evaluated the use of perlite waste in the manufacture of geopolymers by the two parts method was that of Vance et al. (2009), and the compressive strength results for this waste were close to zero, however, when fly ash was added to the mixture, after four days of curing, 37 MPa of compressive strength was obtained. In this study, it was possible to observe a better performance of the geopolymer with perlite waste, close to 1.1 kPa with 7 days of curing, in the stabilization of the iron ore tailings by-product, thus revealing that the reactions and parameters obtained can vary widely according to the amount and type of material, activator concentrations, temperatures and curing times, and, in the case of soil stabilization, with the type of soil as well.

6. Conclusion

The results obtained for unconfined compressive strength indicate that the use of geopolymers in stabilizing iron ore tailings by-products is promising. In addition to providing an appropriate destination for the perlite waste and tailings disposed of in dams, which would be of great benefit in terms of sustainability, it has the potential to be an economic solution as well.

In this paper, two variables were evaluated for geopolymer mixtures in stabilization: the molar concentration of the activator solution and the geopolymer content in the mixtures. The results regarding the unconfined compressive strength showed an increase in this property with increasing variables. Comparing the samples with 20% geopolymer and 80% by-product in the mixtures, an increase in the peak strength from 34,11 to 254,33 kPa (7 times higher) was observed for activator solutions with molar concentrations of 2 and 5 M, respectively. In the samples with 30% geopolymer and 70% by-product, the variation was from 59,16 to 1129,37 kPa (19 times higher). As for the contents, it is possible to see that for the same molar concentration, of 2M, there was an increase in resistance from 34,11 to 59,16 kPa (almost 2 times higher), and for 5M, the difference was from 254,33 to 1129,37 kPa (4 times higher). It should be noted that lower contents of the geopolymer applied in the by-product were previously evaluated in this study. However, the results were not significant, even lower than those obtained for the 2M concentration, indicating that the amount of activator was still insufficient for the geopolymeric reactions to occur.

It is worth mentioning that in all samples a liquid/solid ratio of 0.8 was used and the by-product was in a dry state, the curing time was 7 days, and in the case of precursors, perlite waste and iron ore tailings by-product itself were added as an additional source of silica and alumina.

In future studies it is expected to evaluate other important variables in geopolymer reactions, such as different types of precursors and activators, different curing times, temperature, liquid-solid ratios, and molding moistures, in the case of soil stabilization.

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