Stabilization of iron tailings with alkali activated binders.

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Abstract

Mine tailings are a challenge for the mine activity as they need to be managed and disposed safely. One common solution is the disposal in upstream tailing dams, which have been discouraged and even prohibited in Brazil due to recent accidents with environmental and human loses.

Alternatively, dry stack of filtered tailings are gaining market and many studies are being developed on this subject. Although this solution seems to assure higher safety factors due to the lower water content of the filtered tailings in comparison to the slurry disposed in dams, failures have also occurred on these structures. The dry stack stabilisation can be ensured with a berm of cemented filtered tailings, typically with Portland cement. To reduce the environmental impact, in this work Portland cement (whose production releases a large amount of CO_2) has been replaced by an alkali activated binder (AAB), using fly ash and a sodium hydroxide solution. The use of sodium silicate, very common in AAB, was not considered due to its higher carbon footprint.

This work presents an optimisation study to obtain the best mixture of an iron tailings and the AAB. For this purpose, a statistical approach based on the design of experiments was used considering three main variables: the liquid content, the fly ash content, and the sodium hydroxide concentration. Uniaxial Compression Strength (UCS) tests at 7 days of curing time were performed in specimens moulded in different conditions and the mixture with a higher strength was selected for further analysis. This in-depth study includes the evaluation of UCS and elastic stiffness evolution with curing time. This later was assessed by measuring the shear waves propagation time with ultrasonic piezoelectric transducers. The curing temperature was found to have a significant influence on strength and stiffness evolution.

Introduction

The tailing deposition

An alternative to the disposal in tailing dams is the deposition in dry stacks, where filtered tailings are placed in embankments with much lower water contents than the slurry deposited in dams. In dry stacks, compaction is possible which is an advantage in comparison to the dam, where tailings are deposited in a hydraulic fill. However, these stacks also have their limitations, since the filtered tailings water content is usually higher than the optimum water content of Proctor compaction curve (Davies, 2011). Considering the usual need to achieve very large embankment heights, high effective stresses can be mobilized at the base of the dam and the material is often on the wet side of the critical state line being, therefore, contractile upon shear. As a consequence, the undrained instability observed in liquefaction phenomena is also a concern in these structures. A possible solution under study is the stabilization with cemented berms, made by the mixing the filtered tailings with a binder such as Portland cement (Carvalho *et al.* 2023).

The environmental impact of Cement Portland

The most common binder used for this purpose is the Portland cement. However, its use is currently discouraged due to its carbon footprint. According to data from WBCSD (2002) about 3% of global greenhouse gas emissions and approximately 5% of CO_2 emissions were cement industry responsibility. In addition to the high amount of CO_2 released in the clinker production (40%), there is still a significant amount generated by the production process (50%), by transport (5%) and by the use of electricity (5%). As a consequence, this industry has a high polluting potential. Taking this into account, it is our responsibility to seek for more eco-friendly alternatives and economically viable.

The alternative: alkali activated binders

Considering the need to reduce the environmental impact, an alternative binder has been studied made by the alkali activation of industrial wastes (called herein AAB). AAB are obtained by a chemical reaction between aluminosilicate materials – precursors – with alkali or alkali-based earth – activators – generating polymeric bonds Si – O – Al (Pinheiro *et al*., 2020). The precursors must be rich in silicon and aluminium, so the fly ash fits. Activators originate from alkaline soluble metals, most often based on sodium and potassium (Wallah & Rangan, 2006). This reaction leads to the formation of a gel, which is called C-A-S-H (2D structure) when the calcium content is significant, and N-A-S-H (3D structure) when this content is low (Davidovits, 2005). This gel allows the system reorganization due to increased network connectivity, resulting in a three-dimensional network of aluminosilicate associated with the geopolymer (Duxson *et al.*, 2007).

Some studies have been published regarding the application of AAB to soils (Glendinning *et al.*, 2011; Sukmak *et al.*, 2013; Rios *et al.*, 2017; Rios *et al.*, 2019; Pinheiro *et al.*, 2020) and mine tailings (Xu, 2013; Manjarrez & Zhang, 2018; Obenaus-Emler *et al.*, 2020; and Cristelo *et al.*, 2020).

Statistical model

In order to minimize the amount of laboratory experiments, a statistical plan was carried out known as Response Surface Method (RSM) (Rivera *et al.*, 2019; Pinheiro *et al.*, 2020). The conventional way to obtain the optimized mixture would be to change the amount of each constituent at a time (complete factorial plan), which leads to a high number of specimens to be made and tested. In addition, this method does not allow the identification of interactions between variables. In this work, a composite plan of centered face was used, that is, the axial points are at the center of each face of the factorial space (Anderson & Whitcomb, 2016). This approach has been used not only for the optimized mixture identification, but also for the main variables and outliers' values identification. Three input variables were considered – ash content (A); liquid content (B); and activator concentration (C). The ash content is the ash weight divided by the dry soil weight; the liquid content is obtained by the ratio of liquids divided by the solids of the mixture; and the activator concentration, being, in this case, the concentration of the sodium hydroxide (SH) solution. The range defined for each input variable was 0.1 to 0.3 for (A); 0.14 to 0.17 for (B); and 5 to 11.76 mol/kg for (C). Although a wide research plan is ongoing, in this paper the first phase of the testing plan, comprising 11 specimens, is presented which already shows significant differences between treated and untreated specimens. The main output variable considered for analysis was unconfined compressive strength (UCS).

Methodology

Materials

Iron tailings from Minas Gerais (Brazil) was used in this work, whose grain size distribution curve can be seen in Figure 1. The particles unit weight (γ_S) is quite high (29.5 kN/m^3) when compared to natural soils due to the iron content. The tailings did not present plasticity and liquidity limits, being classified as non-plastic (NP). The precursor used in this work is type F ash, i.e., with low lime content (< 5 %). It has in its composition: SiO_2 (54.84 %), Al_2O_3 (19.46 %), Na_2O (1.65 %) and CaO (4.68 %). Typically, activators are composed by the combination of sodium hydroxide (SH) and sodium silicate (SS). In this study, the use of SS was eliminated since its fabrication process also has a great environmental impact. The SH solution consists of 32 % NaOH and 68 % water, with a concentration of 11.76 mol/kg, which was diluted for the lower concentrations.

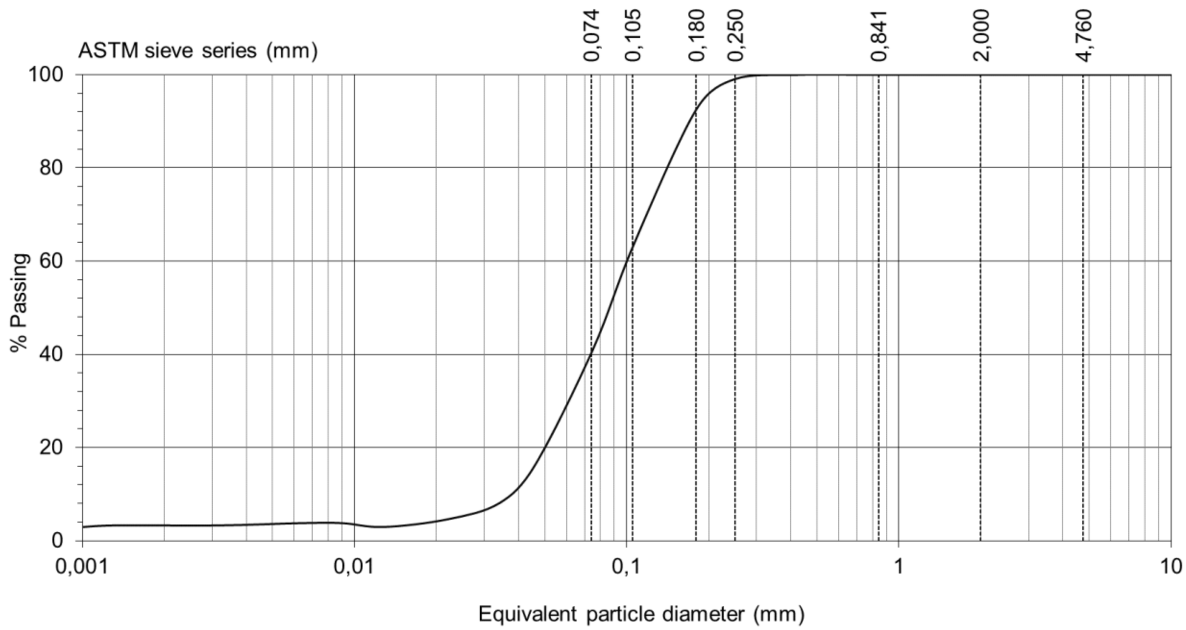


Figure 1: Iron tailing grain size distribution curve

Sample preparation

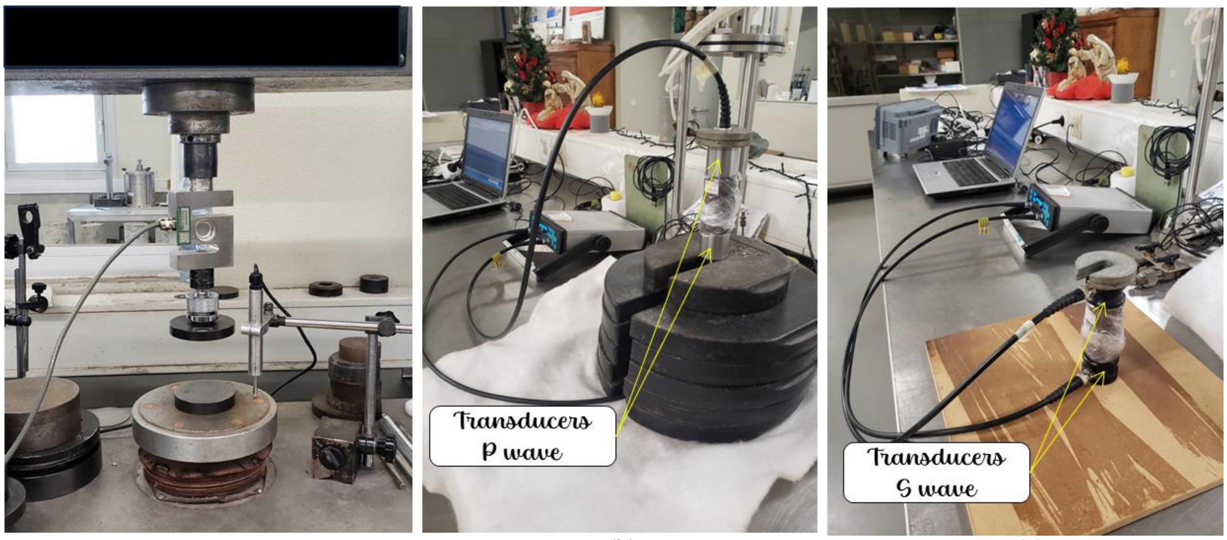
Molds of 38 mm in diameter and 65 mm in height were used for the specimen preparation. The tailings were initially oven dried to allow the moisture control. However, as filtered tailings still have a significant water content a fixed quantity of water was added to the soil. In this case this amount was fixed to 15 ml, which corresponds to a water content between 14 % and 17 % depending on the ash content. Thus, alkali activation was tested for different soil moistures. Because of this water addition, the SH concentration was eventually diluted.

Initially, 15 ml of water was added to the dry tailings to make them homogeneous. Then, the ash was incorporated, and, after homogenization, the SH was added. Once all the components were incorporated, the mixture was transferred into a sealed plastic container to avoid moisture loss. Compaction was performed in three equal layers to obtain the target dry unit weight (γ_d). The specimen extraction was carried out on the same day, being then stored for 7 days.

To study the influence of curing temperature on the specimen's strength and stiffness, some specimens were placed in a low temperature oven for 24 hours. After this time, they were stored at room temperature until the seventh day. At that time the specimens were removed from the box and wrapped in plastic film for better handling and to keep the specimen moisture.

Testing procedures

Unconfined compression strength tests were performed following ASTM D2166-06 (2006) standard for the untreated tests and the Portuguese specification (LNEC E 264, 1972) for the treated specimens, using a load cell of 20 kN of capacity and a constant rate of 0.02 mm/min, using the test setup indicated in Figure 2.a.



(a) (b) (c)

Figure 2: Performed tests configuration: a) UCS test; b) P waves reading; c) S waves reading.

Seismic wave velocities measurements were performed following EN 12504-4 (CEN, 2003), to obtain the propagation time of compression (P) and shear (S) waves towards the calculation of elastic parameters. These measurements were performed with piezoelectric ultrasonic transducers as described by Rios *et al.* (2017) and different input frequencies were used depending on the specimen type and curing time (Viana da Fonseca, Ferreira, e Fahey 2009); Ferreira *et al*., 2021). Readings were performed at 7, 14, 21 and 28 days. The great advantage of this test is to be non-destructive, allowing to do measurements in the same specimen over the 28 days of curing time and at the end the specimen is still suitable for other tests such as a UCS test, for example. The configuration of this test can be seen in Figure 2.b for P waves and Figure 2.c for S waves.

Data and discussion

The selected mixture

After executing the statistical plan, UCS tests were performed at 7 days in the 11 mixtures. The mixtures input variables are shown in **Error! Reference source not found.**, where the parameter A is the fly ash content; B is the liquid content and C is the SH concentration and the UCS result of all mixtures is shown in Figure 3. The mixture that presented the highest strength (ID-8) was selected as optimized mixture. Its input variables are: (A) 0.30; (B) 0.17; and (C) 11.76 mol/kg. The temperature influence on strength and stiffness will be studied only in this mixture.

UCS tests were also performed on the untreated tailings according to the following compaction degrees based on Normal Proctor test – 90 %, 95 % and 100 % and the obtained results are presented in Table 1 (by the horizontal lines). As there is no chemical reaction the strength does not evolve in time and so the specimens were sheared on the molding day. It is noted that the maximum strength (40 kPa) was obtained for the highest compaction degree but is much lower than the strength obtained with the optimum treated mixture (182 kPa). Although the compaction degree of the treated specimens ranged from 90 % to 95 % of Normal Proctor, all these specimens had higher strength than the untreated soil at 100 % of Normal Proctor (with exception of mixture 1 which has slightly smaller values).

Table 1: Input parameters mixtures.

ID	1	2	3	4	5	6	7	8	15	16	21
A	0.10	0.30	0.10	0.30	0.10	0.30	0.10	0.30	0.20	0.20	0.20
B	0.14	0.14	0.17	0.17	0.14	0.14	0.17	0.17	0.16	0.16	0.16
C	5.00	5.00	5.00	5.00	11.76	11.76	11.76	11.76	8.50	8.50	8.50

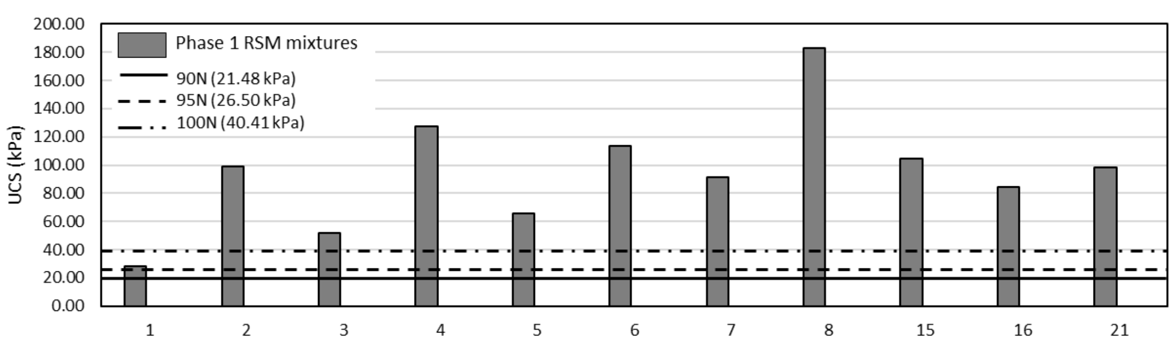


Figure 3: UCS Results.

Effect of curing temperature

Stiffness evolution

Readings of the propagation time for compression (P) and shear (S) waves were performed at 7, 14, 21 and 28 days for the specimens that were 24 hours in the low temperature oven after molding and for the specimens cured at room temperature. The distance between the transducers divided by the wave propagation time gives the wave propagation velocity. From the shear wave velocity (V_S) the maximum shear modulus (G_0) can be determined by equation 1:

$$G_0 = \rho_m \times V_S^2 \tag{1}$$

where ρ_m is the mixture density obtained by the ratio between the specimen weight and its volume. The results of the stiffness evolution over 28 days of curing time in the specimens cured with and without temperature is observed in Figure 4.a. The stiffness of the specimen cured with temperature is already much higher at 7 days than the stiffness at 28 days of the specimen cured without temperature. However, in percentage, the sample without temperature had a stiffness increase of more than 200 % (140 to 432 MPa) while the other sample (with temperature) had a stiffness increase of around 50 % (832 to 1245 MPa). It is also interesting to notice that the evolution shows a tendency to continue increasing over time.

Strength evolution

Unconfined compression strength evolution with curing time was evaluated also for the two groups of specimens (cured with and without temperature) for 7, 14, 21 and 28 days. In mixture without temperature the strength increases gradually over time, totaling an increase at 28 days of 27 % compared to the value obtained at 7 days. The mixture cured with temperature showed a dubious trend. It has a high strength increase from 7 to 14 days, and then it reduces having the value of 28 days lower than that obtained at 21 days, but still higher than the initial and that all the specimens that were not cured with temperature. These curves can be seen in Figure 4.b. A possible explanation for this observed characteristic is associated with a possible temperature variation in the oven, since the 7 and 28 days were molded in one day and those of 14 and 21 was molded in another day when the oven temperature was higher. Further studies are being conducted with greater temperature control to better understand this trend.

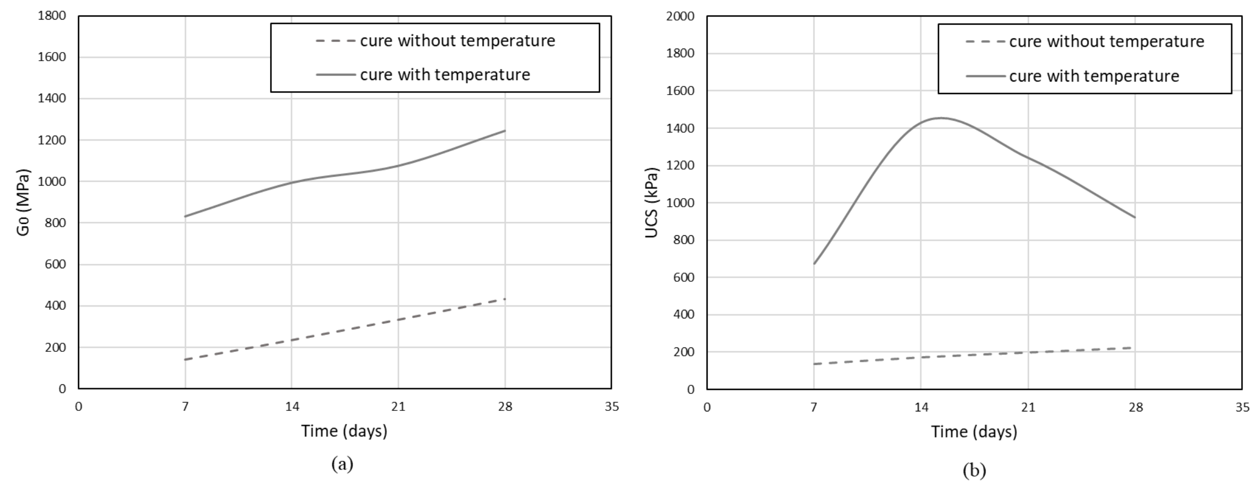


Figure 4: Evolution over 28 days: (a) Stiffness evolution; (b) Strength evolution.

Conclusion

The statistical model (RSM) facilitated the identification of the optimal mixture, since from only 11 specimens it was possible to reach a mixture with satisfactory strength well above the UCS strength obtained in the soil without treatment. Elastic stiffness evolution was almost linear for both cases cured with and without temperature, but the values cured with temperature are much higher indicating that this method is suitable for applications in hot climates. Strength also increased with temperature, although further studies are being conducted with greater temperature control to better define the UCS trend with time.

Acknowledgements

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