The Influence of Aggregate Microstructure on Tailings Behaviour

Jiri Herza 1, Daniel Jirasko 1, Ivan Vanicek 1

1 Department of Geotechnics, Faculty of Civil Engineering, Czech Technical University, Thakurova 7, 166 29 Praha 6 – Dejvice, Czech Republic

Jiri.Herza@fsv.cvut.cz

Abstract. Conventional constitutive soil models applied in geotechnical engineering have been developed using the phenomenological approach from macroscopic observations and application of continuum mechanics. Despite the adopted simplification and homogenisation of complex interactions of soil particles, water and air, the phenomenological models and the underlying principles have been proven to satisfactorily describe the stress-strain conditions in most common soils. However, the application of the conventional soil behaviour models has been proven to be difficult in the mining industry and tailings storage facilities in particular, where the structural integrity of the storage facilities often depends on the performance of extensively altered materials (tailings) that are produced as a waste product from ore processing. The difficulties stem from the altered nature of the tailings and the inconsistencies in the tailings deposition, which may result in the formation of porous and brittle matrices of heterogeneous aggregates. Such matrices are susceptible to static and dynamic liquefaction whereby little changes in the stress-strain conditions can result in a sudden collapse of the soil structure. Sampling and preservation of intact delicate tailings is often unachievable, and the analyses of the laboratory tests are obscured by the simplifications, assumed homogenisation of the sample and the inherent influence of the testing apparatus on the measured properties. Therefore, the characterisation of the in situ conditions of the tailings and the prediction of the tailings performance for altering stress-strain conditions using conventional soil behaviour models is very difficult. Yet, understanding the delicate state of the brittle tailings is crucial for upstream and centrally raised tailings storage facilities. This is exemplified by the facility failures which featured devastating consequences in Brazil in 2015 and 2019. These failures were caused by liquefaction of tailings, which acted as foundations for the upstream raised embankments. This paper discusses the influence of the tailings micro-structure on the macroscopically observed behaviour. This paper also shows that, using modern optical and other testing methods, the micro-structure can be measured and then allowed for in the soil behaviour model with the aim to improve the reliability of tailings liquefaction assessment.

1. Introduction
Shear resistance of soils have significant direct and indirect impacts on many factors governing the design, construction and performance of geotechnical and civil structures. These factors include slope stability, bearing capacity, penetration resistance, foundation deformation and earth pressure acting on structures. Due to its importance, the shear resistance of soils has been extensively studied for over 200 years. Since the 18th century, the stresses acting in the normal direction to a shear surface have been understood to control the shear strength of soils. The original concept, presented by Coulomb in 1773...
was advanced by the effective stress theory introduced by Karl Terzaghi in the mid-1930s [2]. Further research during the 20th century led to a greater understanding of the underlying principles and the development of the Critical State Soil Mechanics [3] and Steady-State theory [4], both of which are used today.

Shearing of soils is a complex, multiphase process. It involves sliding of particles along each other, rotation of particles, displacement of the particles within the soil matrix and redistribution of water and air in the void between the particles [5]. Shear resistance is generated by the normal stress through engagement of particles (aggregates) within the matrix of the sheared soil. Given that soil is not a rigid body, loading of the soil (to generate normal stress) is accompanied by deformation (strain). At low strains, the shear resistance of the soil is activated, as an increasing number of particles are engaged in the multi-phase process. As the shearing progresses, the particles’ position, orientation, and even composition change and heat is generated. If water is present then water pressures are generated and dissipated, resulting in continuously changing internal effective stresses and assembly of aggregates within the sheared soil.

The constitutive soil models applied in geotechnical engineering have been developed based on a phenomenological approach from the macroscopic observations (experiments) and application of continuum mechanics. The analogy between soil and continuum stems from the constitutive relations between the stresses and geometry of the sheared soils that are inferred from laboratory tests. The forces or stresses and displacements measured at the soil mass boundaries from laboratory tests are postulated to be distributed over the whole volume of the soils at a respective scale. This homogenisation ignores the particulate nature of the material, even though the macroscopically observed response is often an aggregation of the complex patterns of local discontinuities.

The key attributes that control the soil behaviour are the applied stresses and the soil structure, which is influenced by many factors such as mineralogy, stress history, void ratio etc. Casagrande [6] observed that the terminal stage of shearing of granular materials eventually reached a volumetrically stable condition when sheared under the same normal stress. In this state, later termed Critical State, the soil volume and shear stress remain steadily constant along with the distortion of the sample.

Noting the uniqueness of the terminal stages of shear tests, Wroth [7] further developed the initial Critical State theory by Casagrande and postulated that the final void ratio (representing the soil structure in the context of this paper) and shear stresses are related to the applied normal effective stress and are independent of the stress history. Both Casagrande and Wroth described the phenomenon observed in a simple shear apparatus, where normal and shear stresses acting on the failure plane are given, while the stress components out of this plane are unknown. Roscoe et al. [3] extended the earlier Critical State concept to apply to triaxial compression using tests where the stress components acting on each plane were completely known. To generalise the definition of the Critical State, normal and shear stresses were then replaced by the mean (confining) effective stress ($p'$) and the deviator stress ($q$) invariants.

Figure 1 shows the Critical State Locus (CSL), which refers to a curve in the $p'$- q - e space connecting the points of balance (points a, b and c) between the internal structure (expressed by the void ratio, e) and the deviator ($q$) and mean effective stresses ($p'$). At these points, a continuous constant rate of shearing is possible without changes in the effective confining and deviator stress. Figure 1 was composed from idealised triaxial compression stress paths of a clean quartz sand and it also shows the projection of the CSL onto the $p'$-e and $p'$-q planes.
Figure 1. Locus of the critical state in \( p' - q - e \) diagram

CSL was initially developed using the Cam-Clay model, which assumed that the soil was isotropic, elasto-plastic and it deformed as a continuum not affected by creep. As natural soils violate the basic Cam-Clay assumptions, the concept has been modified by various authors (e.g. Jefferies and Been,[8]) to be more applicable. The concept of Critical State Soil Mechanics (CSSM) has increasingly become a popular tool for interpreting the mechanics of granular materials, having been validated by numerous researchers and studies.

The CSL model was also adopted in constitutive soil models such as NorSand that builds upon the fundamental dependency of the soil response on the present void ratio and stresses. Following the failure of tailings Samarco and Funjao dams in Brazil, the CSSM concept has become the preferred method of describing a tailings liquefaction potential and risk.

The vast majority of laboratory tests to define the shear resistance of soils and indeed the CSL are conducted in triaxial compression and CSSM assumes that the unique CSL is reached irrespective of the stress path as shown in Figure 1. However, several authors ([9], [10] and [11]) have observed that the CSL was not unique and different results were produced by triaxial compression and extension tests. It was also indicated that the CSL for a given material differs for different initial stress levels. From these observations it appears that the stress history may not be fully “forgotten” by the shearing process at large strains and that both the initial soil fabrics and loading mode influences the shape of the CLS.

Irrespective of the CSL’s uniqueness or the validity of CSSM, a soil’s state parameters \( p' \), \( q \) and \( e \) and their derivatives play fundamental roles in calculating engineering properties of soils. The state parameters (discussed later) are also used to assess the potential of a soil structure to collapse or liquefy [12], [13], [14] and [15].

This paper discusses the potential impact of the soil microfabric and composition on the engineering properties using microscopic investigations methods and the relevance of the void ratio for description of the microscopical processes in soils. The soil microstructure and complexity is shown on samples of bauxite residue from bauxite operations in Spain (sample ALSP) and Western Australia (sample
and a reference sample of a river quartz-based sand (RESN). The choice of bauxite residue was driven by the unique nature of the residue [16] and because the engineering properties of the bauxite residue are fundamental to the safety of several very large residue storage areas across the globe especially where the upstream construction method was used [17].

2. Bauxite residue
Alumina, which itself is smelted to produce aluminium metal, is produced from bauxite by means of digestion and filtration, which is commonly referred to as the Bayer process (after Carl Josef Bayer). Bauxites contain alumina mainly in the form of gibbsite (Al(OH)_3), boehmite (AlOOH) and diaspore (α-AlO(OH)). To extract the alumina the raw bauxite ore is crushed and milled and then digested in pressurized vessels with sodium hydroxide at temperatures from 150°C to 270°C. In these conditions, most of the alumina rich minerals are dissolved. The insoluble residue is removed from the solution by settling and filtration.

The coarser fraction (residue sand or red sand) and the finer fraction (red mud) of the bauxite residue is then separated typically by means of hydrocyclones. The separated red mud is further treated (thickened and sometimes filtrated) before being disposed of in Residue Storage Areas (RSA). The red sand is often used for construction of drainage layers and embankments containing the red mud in the RSAs.

The red mud leaves the Bayer process as a highly alkaline slurry with pH of over 10 and it typically contains particles of silica, aluminium, iron, calcium and titanium oxides and hydroxides. Depending on the bauxite source, the red mud can be toxic because of the sodium hydroxide and calcium complexes. The RSAs are therefore considered as hazardous facilities, failure of which can have devastating impact on the surroundings as demonstrated by the Ajka dam failure in October 2010, which claimed 10 lives and cause significant environmental and economic damage [18]. WISE-uranium (wise.uranium.org) lists another five failures of bauxite RSAs since 2007.

While the red mud material has been the subject of numerous researches over the last decades, the residue sands have been left aside. This is perhaps due to the red mud forming the majority of the bauxite residue mass and has traditionally been considered, from a geotechnical and engineering perspective, to be more critical than the comparatively coarser sand. This perception stems from the finer nature, lower permeability and lower shear resistance of the red mud than compared with the residue sand.

The residue samples for this research was acquired from conventional bauxite operations in Spain (sample ALSP) and Western Australia (sample ALWA). Both operations use the Bayer process and the sand residue was separated from the bauxite residue stream using hydrocyclones.

3. Residue microstructure and mechanical performance
3.1. SEM analysis
The initial assessment was completed via a conventional Scanning Electron Microscope (SEM) with an Energy Dispersive Spectroscope (EDS). The chemical composition of the samples was confirmed using X-Ray Diffraction technique.

The SEM images of the residue sample at the minimum magnification (310x) clearly indicated the vast difference between the residue sand and the reference sand (RESN). As shown in Figure 2, the residue sand aggregates are bonded together from smaller particles, while the sand grains are formed by compact masses of solid matter.
The EDS analyses indicated that the ALSP sample comprises mainly aluminium and iron oxides and hydroxides, with minor presence of titanium and sodium oxides. The oxides, most commonly Goethite, Hematite and Gibbsite were found to be bonded together to form very porous grains ranging in size from units of microns (Figure 3, left) to hundreds of microns (Figure 2 – centre). The larger aggregates were also found to be bonded by oxide/hydroxide bridges to form even bigger complexes (Figure 3, right). The structure of the residue sand is similar to the structure of laterite soils in Western Australia as discussed in Herza and Terzaghi [19]. This similarity is not surprising because the bauxite ore was formed by in-situ weathering (laterization) of the rock units.

The topology of the bonded aggregates were very complex and the surfaces were rough on a microscopic level as shown in Figure 3 – left.

The ALWA sample contained mainly quartz, Gibbsite, Hematite and Goethite. The quartz was present within aggregates bonded together (Figure 4, left) and as individual grains (Figure 4, centre).
Figure 4. SEM images of ALWA aggregates

The quartz grains in the ALWA samples had very complex surfaces with cavities and grooves (Figure 4, right) likely caused by the mechanical and chemical alteration of the aggregates in the Bayer process.

3.2. XRD

The results from the X-ray powder diffraction (XRD) test (Table 1) completed on pulverised samples confirmed the preliminary composition of the material indicated by SEM and EDS.

| Name of mineral | Chemical formula | % content by weight in ALSP sample | % content by weight in ALWA sample | Specific gravity of minerals |
|----------------|------------------|------------------------------------|-----------------------------------|----------------------------|
| Goethite       | FeO(OH)          | 52.8                               | 17.9                              | 3.80                       |
| Hematite       | Fe₂O₃            | 21.6                               | 14.6                              | 5.20                       |
| Gibbsite       | Al(OH)₃          | 13.2                               | 20.2                              | 2.34                       |
| Boehmite       | AlO(OH)          | 12.4                               | 0.7                               | 3.03                       |
| Quartz         | SiO₂             | 0.0                                | 43.4                              | 2.65                       |

3.3. Porosimeter test

The porosimeter test was completed in a mercury porosimeter on loose samples. If the samples were in denser states, the distribution of the void sizes would be different. The method only detects voids with maximum size of 100 μm and hence the relatively large spread of the loosely packed sample aggregates does not significantly impact the results.

As can be seen in Figure 5, a large proportion of the porosity is formed by voids with a pore size smaller than 1 μm. Given the sandy nature of the tested residue samples and based on the SEM images, it appears that the sub 1 μm voids are the voids within the bonded aggregates. From this perspective the residue samples have dual porosity being the intra-aggregate porosity and the inter-aggregate porosity and the same applies to the void ratio. While the inter-aggregate void ratio (e) is the ratio that is used to represent the material structure in the p’- q - e space (Figure 1) the intra-aggregate void ratio is inert and does not take part in the volumetric changes of the material under loading (consolidation, shearing, etc.). This secondary, intra-aggregate void ratio does not exist in conventional coarse grained materials.
3.4. Nanoindentation test
An instrumented indentation technique was used to estimate the elasticity modulus (E) of goethite and hematite aggregates and 44 micro dents were made. The resistance of the target aggregates was found to be similar to the resistance of hydrated cement. The modulus of elasticity was calculated to vary between 23 and 33 GPa, which corresponds to the values presented by Chicot et al [20]. For comparison the E values of pure goethite and hematite are an order of magnitude greater (> 500 GPa), and pure quartz has an E close to 100 GPa, while E of common sands is in the order of tens of MPa.

Given that the elasticity modulus of the bonded aggregates was found to be 100 times greater than the elasticity modulus of sands, the structural integrity of the bonded aggregates is unlikely to be compromised under any loading conditions conceivable for geotechnical structures.

The strength of the oxide and hydroxide interaggregate bridges (Figure 3, right) is planned to be tested in the next phase of the research. However, as the simple reworking of the residue samples in a mortar grinder resulted in significant changes of the grain size distribution (discussed later), the shear strength of the bridges is expected to be much smaller than the strength of the bonded aggregates themselves and these bridges are likely to break during shearing.

3.5. Geotechnical classification tests
The samples were classified using conventional particle size distribution (PSD) and Atterberg Limit tests and specific gravity test. The results of the PSD are plotted Figure 6. The Atterberg Limits test could not be completed as all samples were non-plastic. Based on the classification testing the residue sands and the reference sand samples were classified as poorly graded medium to fine sands (SP) in accordance with ASTM.

To estimate the impact of the internal structure on the particle sizes, the PSD test was repeated on remoulded samples (worked by shearing in a mortar grinder for 30 seconds). The results indicated that remoulding the residue samples made them finer with the proportion of fines increasing by up to 12 % (Figure 6). Remoulding of the RESN sample did not have a noticeable impact on the sample PSD. The PSD results indicated that the shearing of the residual sands could breakdown the inter-aggregate bonds with further impacts on the material performance.
Figure 6. PSD plots

A specific gravity (SG) of the samples was measured in a helium pycnometer and it returned values of 2.67, 3.44 and 3.38 for the RESN, ALSP and ALWA, respectively.

As shown in Table 1, the difference between the minerals that form the residue aggregates is as much as 100%. The SEM images showed that the distribution of the various minerals is not homogeneous and thus the material SG can be spatially variable. In addition, the higher SG aggregates can quickly segregate from the lighter materials and the PSD tests indicated potential breakage of the interaggregate bonds. Therefore, depending on the technique used for deposition of the residue sand, the residue sand can be highly variable in terms of its SG, void ratio and materials composition within the RSA structures.

3.6. Shear strength tests

The initial shear strength tests were completed in a ringshear apparatus with an external and internal diameter of 100 mm and 50 mm, respectively and a strain rate control. The samples were tested in saturated drained conditions after being consolidated to 50 kPa.

The test results of the RESN sample returned a typical stress-strain curve and an expected friction angle of approximately 36 degrees and no cohesion. The tests results of the residue sand indicated a friction angle of 45 degrees and an apparent cohesion of 10 kPa. The indicated cohesion is most likely the result of particles interlocking and it diminished when the residue sands were later retested at greater consolidation stresses.

Although the residue sand samples were sheared to more than 100 mm strain the expected balance between the shear stress, effective normal stress and sample volume (that is, the aforementioned Critical State) was not reached and the shear resistance of the samples decreased as the sample was further sheared. This indicated that the continuous shearing gradually led to a breakdown of the bonded aggregates into smaller grains. These smaller grains then take part in the stress redistribution and the ‘active’ void ratio of the material increases throughout the shearing process. The shear test results also indicated that the residue sand may exhibit brittle behaviour where a rapid increase in shear resistance is followed by a rapid drop.
It should be noted that the detail assessment of the shear tests completed is outside the main focus of this paper and further testing, captured in a dedicated publication will be completed as part of the next stage of this research.

4. Discussion and conclusion
Samples of bauxite residue were tested together with reference samples of a river-type quartz sand. Based on the geotechnical classification tests, the residue materials and the reference sample were identically classified as poorly graded sands with similar PSDs.

The mineralogical and SEM analyses identified that the ALSP residue sample did not contain any quartz and the ALWA sample contained only about 43% of quartz grains by weight. The microscopic assessment of the residue structure confirmed that the majority of the ALWA sample and the entire ALSP sample comprised iron and aluminium oxides and hydroxides, which were bonded together to form a very porous, sand-size aggregates. These aggregates were then bonded together by oxide and hydroxide bridges to form yet larger complexes.

The nanoindentation probing indicated that the elasticity modulus and hardness of the bonded aggregates was in the same order of magnitude as was quartz. Because the elasticity modulus of sands and other soils were two orders of magnitude smaller than the elasticity modulus of the bonded aggregates, it was unlikely that loading of the soil (in compression or shear) would cause complete destruction of the aggregates. Therefore, a very large proportion of the measured voids would not take part in the volumetric changes during shearing and would remain passive in respect to the interaction between the effective confining and deviator stresses.

However, the PSD tests before and after the residue remoulding and the shear strength tests indicated that the bridges between the bonded aggregates can be easily broken and the initial shear resistance of the tested samples was reducing with increasing strain. It appears that the continuous shearing gradually breaks down the bonded aggregates into smaller grains, which take part in the stress redistribution and the ‘active’ void ratio of the residue sand may increase throughout the shearing. Because the void ratio of the residue sand comprises both ‘passive’ and ‘active’ voids and because the balance between these two groups may be changing during shearing and other deformations, the void ratio may not be the best indicator of the residue state for constitutive geotechnical models, as is used in currently accepted soil mechanics theories.

Advanced microscopic investigation and testing method significantly assist with understanding the complex structure of the residue sands and the impact on the soil performance. These methods combined with conventional performance testing have the potential to identify a more suitable parameter(s) to capture soil structure. This parameter(s) could then be applied in soil behaviour models to define the current state and future states of residue, tailings storage facilities and other geotechnical structures.

Acknowledgment
This work was supported by the Grant Agency of the Czech Technical University in Prague, grant No. SGS20/044/0HK1/1T/11.

References
[1] C. A. Coulomb, “Essai sur une application des regeles de maxims et minimis aquelques problemes de statique, relatifs a l’architecture;” Me’moires de Mathe’matique et de Physique, 7, 343–382, 1773.
[2] K. Terzaghi, “A fundamental fallacy in Earth pressure computations,” Journal of the Boston Society of Civil Engineers, 23, 13–32, 1936.
[3] K. H. Roscoe, A.N. Schofield and C. P. Wroth. “On the yielding of soils,” Géotechnique, 8,
22–53, 1958.

[4] S. J. Poulos, S.J. “The Steady State of Deformation,” Journal of Geotechnical Engineering, Div, Am, Society Civil, 107, 553-562, 1981.

[5] J. K. Mitchell, K. Soga. “Fundamentals of Soil Behavior”, 3rd Edition. ISBN: 978-0-471-46302-3, 2005.

[6] A. Casagrande, “Characteristics of cohesionless soils affecting the stability of slopes and earth fills,” Journal of the Boston Society of Civil Engineers, 23, 13–32, 1936.

[7] C. P. Wroth, “Shear Behaviour of Soils,” Ph.D. Thesis, Cambridge University, 1958.

[8] M.G. Jefferies and K. Been, “Soil Liquefaction – A critical state approach,” ISBN 0-419-16170-8, 2016.

[9] Y. P. Vaid, J. C. Chern and H. Tumi, “Confining pressure, grain angularity, and liquefaction,” ASCE Journal of Geotechnical Engineering, 111(10): 1229–1235, 1985.

[10] M. F. Riemer and R. B. Seed, “Factors affecting apparent position of the steady state line,” Journal of Geotechnical Engineering, ASCE, 123(3):281–288, 1997.

[11] P. G. Joseph, “Dynamical system-based soil mechanics,” ISBN: 978113872322, 2017.

[12] P.K. Robertson, “Cone Penetration Test (CPT)-based soil behavior type (SBT) classification system – an update.” Canadian Geotechnical Journal, 53(12), 2016.

[13] D. Reid, “Estimating slope of critical state line from cone penetration tests – an update,” Canadian Geotechnical Journal, 52: 46-57, 2014.

[14] S. J. Poulos, G. Castro, and W. France, “Liquefaction evaluation procedure,” Journal of Geotechnical Engineering Division, ASCE, 111(6): 772–792, 1985.

[15] P.K. Robertson, “Evaluation of flow liquefaction and liquefied strength using the Cone Penetration Test,” Journal of Geotechnical & Geoenvironmental Engineering, 136, 842-853.

[16] T. Newson, T. Dyer, Ch. Adam and S. Sharp, “Effect of Structure on the Geotechnical Properties of Bauxite Residue,” Journal of Geotechnical and Geoenvironmental Engineering,132(2) 143-151, 2006.

[17] I. Vanicek, M. Vanicek, “Earth Structures in Transpor, Water and Environmental Engineering”. Springer, 2008.

[18] D. Tury, J. Pusztai and I. Nyari “Causes and Circumstances of Red Mud Reservoir Dam Failure In 2010 at MAL Zrt Factory Site in Ajka, Hungary,” Seventh International Conference on Case Histories in Geotechnical Engineering, 10, Paper No. 3.14a, 2013.

[19] J. Herza and S. Terzaghi “Investigations into Lateritic Soils at Millstream Dam”, 3rd International Conference on Problematic Soils, pp155-162, 2010.

[20] D.Chicot, J.Mendoza, A.Zaoui, G.Louis, V.Lepingle, F.Roulet and J.Lesage, “Mechanical properties of magnetite (Fe3O4), hematite (α-Fe2O3) and goethite (α-FeO·OH) by instrumented indentation and molecular dynamics analysis,” Materials Chemistry and Physics, “129(3), 862-870, 2011.