Clay micromechanics: experimental challenges and perspectives

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ABSTRACT

The importance of physico-chemical processes at the particle scale for the engineering scale behaviour of fine-grained geomaterials is undisputed. Yet, despite great advances in the discipline, experimental evidence that fully resolves the clay micromechanics i.e. linking the evolving microstructure and interparticle actions under loading, is lacking. This paper will discuss the challenges ahead in quantifying the evolving kinematics and interparticle interactions of fine-grained geomaterials. As such, the current limitations, and the potential opportunities of experimental methodologies for manipulating, monitoring and (post-mortem) analysing fine-grained materials at the particle scale will be discussed. In addition to the need of integrating multiple experimental techniques that span several length scales and modalities, the critical role of advanced data reduction and analysis is highlighted, as required for a measurement as opposed to qualitative observation. Throughout the paper, the link between experimental clay micromechanics and modelling will be discussed.

Keywords: Clay, micromechanics, experimental methods

1. Introduction

The behaviour of (natural) clayey geomaterials at the engineering scale (metres) is controlled by the interactions between particles and the evolving microstructure (nm - mm). Understanding such evolution is a critical step in soil mechanics as it allows to interpret experimental data, understand fundamental mechanisms of soil behaviour, develop discrete element models, inform "continuum" constitutive models and eventually design the geotechnical structure.

Clay can be defined either by its size or by the class of mineral. In terms of size, the engineering classification consider as clays all constituents of soils smaller than 2 μm. This definition is justified by the fact that, in soils, this grain size class contains a high fraction of clay minerals, which is often governs the soil properties at the macroscopic level (swelling, shrinkage etc.). In mineralogical terms, clay minerals are primarily phyllosilicates and differ from the other minerals by their small particle size, electrically charged surfaces, plasticity when mixed with water and high weathering resistance (Mitchell and Soga, 2005).

In the case of granular materials (particles ranging from few tenths of micrometres to millimetres), significant advances have been made in understanding and describing the evolution of the interactions at the particle scale. Techniques such as X-ray Computed Tomography (XCT) are combined with advanced image processing methods to measure the evolution of different microstructural aspects of such materials, e.g. shear strain localisation, friction mechanisms and crushing (Oda and Kazama, 1998, Oda et al., 1998, Andò et al., 2012, Zhao et al., 2015). In turn, these experiments have been pivotal to the development and validation of discrete element models to simulate the complex response of granular materials.

Yet, despite great advances in the discipline, experimental evidence that fully resolves the clay micromechanics, i.e. linking the evolving microstructure and interparticle actions under loading, is lacking. Whereas granular materials (or nonclay minerals) are composed primarily of bulky (3D) particles, clay minerals are (2D) platy, and sometimes (1D) needle shaped or tubular, which makes them extremely difficult to image.

Research on the micromechanics of clayey geomaterials is lagging due to the difficulty of investigating particle-to-particle interactions in wet samples at such a small scale. Even very basic responses in 1D/isotropic compression such as reversible ('elastic') and non-reversible ('plastic') behaviours have never been directly observed experimentally.

This paper discusses the challenges ahead in realising the ultimate goal of quantifying the evolving kinematics and interparticle interactions of fine-grained geomaterials in their hydrated state. It will show how the geotechnical community is moving toward clay microstructural analyses that are non-destructive, 3D and multiscale

2. Clay Micromechanics

To fully investigate the soil micromechanics three different aspects need to be described (modified after Calvetti et al. (1997):

- The microstructure, such as the geometrical description of particles position, pore distribution and contacts.
- The kinematics evolution, such as displacements, rotations and the evolution of contacts.
- The interparticle forces, which can be hydro-mechanical and/or electrochemical.

Despite the difficulties of imaging the microstructure of different clay minerals at different environmental condition or stress history, including the sample preparation and technological limits, there is a large amount of work in the literature, starting from the second half of the last century. By using mainly Scanning Electron Microscopy, different particle conceptual models, such as honeycomb, matrix/cardhouse, aggregated and or dispersed structures, were proposed (Lambe, 1958, Sides and Barden, 1971, Collins and McGown, 1974, Van Olphen, 1977, Mitchell, 1956, Sloane and Kell, 1966, Yoshinaka and Kazama, 1973). Recently, such conceptual models were corroborated or refined by investigating the pore space and its distribution by means of Mercury Intrusion Porosimetry (Alonso et al., 1987, Delage et al., 1996, Delage and Lefebvre, 1984, Romero et al., 1999, Monroy et al., 2010, Pedrotti and Tarantino, 2018).

On the other hand, investigations on evolving clay microstructure is lagging behind. The limiting factor in characterising the kinematic evolution of the microstructure of clays has been the lack of in-situ and non-destructive techniques, able to operate at the clay particle scale.

Understanding and measuring clay interparticle forces is extremely difficult and often returns results with very low repeatability (Sposito, 1984, Sposito, 1998, Ganor et al., 1995, Brady and Weil, 1996, Braggs et al., 1994, Rand and Melton, 1977, Rand et al., 1980, Van Olphen, 1977, Mitchell and Soga, 2005). Experimentally, it is extremely difficult to electrochemically characterise the clay particle surface and hence measure the associated interparticle forces. Atomic Force Microscopy appears to be the most promising technique to enable direct measurement of the forces existing between two surfaces (Bickmore et al., 1999, Kumar et al., 2016, Gupta and Miller, 2010, Israelachvili, 2011). However, the difficulty in producing clean data and interpreting them in a reliable way has prevented an extensive use of the instrument for clays. Thanks to the increasing computational capability, numerical methods such as Molecular Dynamics (Alder and Wainwright, 1959, Fenkel and Smit, 2002) and Discrete Element Models (Cundall and Strack, 1979, Anandarajah, 2000), are strongly coming into play as a means of interpretation and validation of the particle-based experiments.

3. Scale of Interest

Depending on the soil type and the macroscopical behaviour that is under consideration, the scale at which the microstructural analysis is to be carried out changes by several orders of magnitude.

In granular materials, the scale of the problem is generally defined by the grain size. Clayey materials, on the other hand, often exhibit a complex, multi-scale structure, with different physical processes controlling the behaviour of the different structural levels.

In Figure 1, a diagram of the possible scales of the problem is reported both in terms of the solid phase (i.e. particles) and in terms of voids (i.e. pores).

The layered structure of clays is based on the combination of two basic crystal structural units, namely the tetrahedral silica sheet and the octahedral alumina sheet. Tetrahedral and octahedral sheets bond together to form layers.

A fundamental characteristic of clay minerals is their electronegativity. The surface of phyllosilicates is indeed not electrically neutral and surface charges exist. These charges have two different origins, namely isomorphous substitutions and local charges. Isomorphous substitutions refer to the substitution of one atom of higher valence with another of lower valence in the clay lattice, without significant change in the layer structure. An imbalanced charge also occurs at the layer end due to interrupted covalent bonds. This charge is amphoteric (its sign changes depending on the pore fluid chemistry) and it is positive at low and neutral pHs, while it is negative at high pHs (Van Olphen, 1977, Palomino and Santamarina, 2005).

The type of layers and the nature of the interlayer bond determine the properties of the different clay minerals. Clays are divided into two main groups, 1:1 structure and 2:1 structure. In the former group, kaolinite is the most common clay. The mineral is formed by unit layers of alternating aluminium octahedra and silica tetrahedra. The basal spacing is constant as water cannot intrude (e.g. 7.37 Å for kaolinite). Particles are hexagonal in shape, with an apothem less than a micron and 10s-100s of nm in thickness. Smectites and illite belong instead to the 2:1 group. Their unit layers are composed by an octahedron aluminium/magnesium sheet sandwiched between two silica tetrahedra. The distance between the unit layers is not constant, ranging from 9.6 Å to complete separation, as water intrudes and hydrates the inter-layer space.

Finally, clay layers stack together to form particles. Depending on the nature of the interlayer and the water content, the number of layers in a particle may vary from

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a few to several hundred (Saiyouri et al., 2004). In the case of smectites and illite, particles are not easily defined, and their size and morphology might change upon hydro-thermal-chemo-mechanical perturbations (Behnsen and Faulkner, 2013, Morrow et al., 2017). The interlayer/interparticle spaces play a fundamental role in the macroscopic behaviour of such clays. They are often undulated, and their dimensions can vary from 100s of nm to several micrometres. The particle thickness can be as thin as 3 nm.

Differences during formation are known to trigger complex structures of particles configuration, where different particles tend to group. Such groups of particles are generally referred to as aggregates/floccules/clusters. When they are present, aggregates are considered as the elementary units controlling some or most of the macroscopic behaviour. Mechanical swelling, for instance, is observed at the macroscopic level but governed by the interlayer properties. Aggregate size and interaction have been recognised to evolve with mechanical and environmental loads (e.g. Tarantino & De Col, 2008, Romero, 2013, Musso et al., 2013). Aggregates can range from few micrometres up to several hundreds of micrometres. When aggregate-like structures are present, the pore space is characterised in terms of intra- and inter-aggregate porosity to indicate the space between particles and between aggregates respectively. Intra-aggregate porosity is generally in the order of 100s of nanometres, whereas inter-aggregate porosity is of 10s of micrometres. Higher levels of particles association can be seen in clays with a particular geological or chemical history (i.e. bonding).

Finally, hydromechanical failure mechanisms are associated to the creation of cracks and fissures that can be as large as a millimetre and as small as several micrometres (Wei et al., 2016).

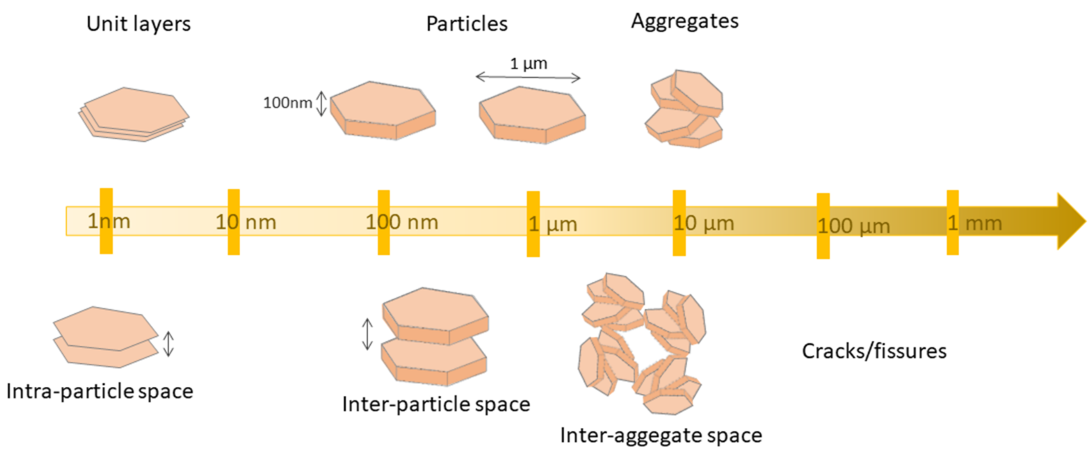

Figure 1. Scale of interest for clay micromechanics

Available measurement techniques, processes, and limitations

Table 1 summarises the most used techniques for the microstructural investigation of clayey materials. Until recently, coupling Mercury Intrusion Porosimeter (MIP), providing the pore size distribution measurement, and Scanning Electron Microscopy (SEM), providing sample imaging and particle configuration, was considered the state of the art for the microstructural characterisation of clays (Hattab et al., 2013).

However, both techniques require heavy sample preparation (e.g. freeze drying) and are post-mortem. MIP data do not provide a spatial distribution and morphology of the pores, whereas SEM allows only for 2D images of an external layer of material.

When the size of pores is below 10nm (beyond MIP capability), gas adsorption can be used as alternative, or as complementary investigation to MIP. It has been intensively used in the characterisation of shales. Despite the different ranges of measurement, gas adsorption presents issues similar to MIP.

Cryo-scanning has recently been adopted to bypass the "freeze-drying" stage in SEM sample preparation. Wet samples are frozen in a controlled environment at high pressure, and imaging is performed without the necessity to lyophilise.

Environmental Scanning Electron Microscopy (ESEM) is an improvement of the traditional SEM, which enables examining wet samples and preserving their natural characteristics (Danilatos, 1993). The examination of the sample can be continuously done at different vapour pressures and temperatures, hence at different relative humidity, making it a suitable method to study the gradual effects of wetting and drying stages at microstructural scale (Airò Farulla et al., 2010). At high water contents, the contrast of ESEM is negatively impacted (Houghton and Donald, 2008), limiting is use mainly to partially saturated clay at low water content.

Table 1. Experimental Techniques

Technique	Sample Size	Destructive (D)/ Non-Destructive (ND)	Sample State	Measure	Inferred Descriptors	Resolution -FoV	Limitation	References
Gas Adsorption	Several grams	D	Dehydrated (freeze-drying/ vacuum)	Adsorbed mass of condensed gas as a function of pressure	Cumulative distribution of the pore volume, specific surface		Sample preparation/ pore morphology and connectivity	Santamarina et al. (2002)
Mercury Intrusion Porosimeter	ca 1g	D	Dehydrated (freeze-drying/oven)	Intruded and extruded volumes of mercury as a function of pressure	Cumulative distribution of the pore volume	3nm – 1mm (pore size)	Sample preparation/ pore morphology and connectivity	Delage and Lefebvre (1984), Romero and Simms (2008), Yuan et al. (2019)
Scanning Electron Microscopy	ca 1g	D	Dehydrated (freeze-drying/oven)	Detection of secondary electrons	Image of the sample surface (with a certain thickness of influence)	1nm – 1mm	Sample preparation, essentially 2D	Romero and Simms (2008), Hattab and Fleureau (2010)
Environmental Electron Scanning Microscopy	ca. 1 g	D	Natural State	Detection of secondary electrons	Image of the sample surface	1nm – 1mm	2D	Romero and Simms (2008), Danilatos (1993)
Cryo-Scanning Electron Microscopy	ca 1g	D	Frozen	Detection of secondary electrons	Image of the sample surface	1nm – 1mm	Sample preparation, 2D	
Confocal Laser/ Electron Microscopy	50 mm	ND	Natural State	Detection of laser or electron beam	Image of the sample surface with increase depth penetration (~3D)	100s nm		Ivashchenko (2022)
Nuclear Magnetic Resonance Spectroscopy		ND	Hydrated	Spectroscopy/ Imaging	Pore Distribution			
X-ray Computed Tomography		ND		Imaging	3D reconstructed volume	~1 μm (lab) < 25-100 nm (synchrotron)	Resolution/ Sample size for nano-tomography (100-200 μm)	
SANS and SAXS		ND	Natural	X-ray/Neutron scattering	Particle spacing and global particle configuration	< 1 nm – 2 μm, ~500 μm	Spatial distribution, large features > μm not resolved, data interpretation	Birmpilis et al. (2019)
Optical Microscope/ polarizing Microscope	50 mm	D	Drying/ Binding agent Impregnation (Carbowax)	Polarised light microscopy	Image of the thin section	~1μm	Resolution/ sample preparation	Morgenstern and Tchalenko (1967)

In the attempt to move from a 2D image to a 3D image, different techniques have been developed and adopted. Broad Ion Beaming/Focused Ion Beaming are techniques that allow to cut and expose thin cross-section of the specimen to be imaged via SEM. Images of subsequent sections of the same specimen allow for a 3D representation of small parts of the specimen (Desbois et al., 2017b). Such techniques still require a post-mortem analysis and prevent an in-situ investigation of the evolution of the micromechanics.

X-ray Computed Tomography (XCT) marked a revolution in the study of the microstructure of granular materials, as it is not destructive, no sample preparation is required, it allows for in-situ analyses and provides a reconstructed 3D image of the sample. Nevertheless, XCT resolution (as high as 25 nm/voxel at synchrotron based nano-tomography instrument) is not yet sufficient to resolve clay particles and the associated pores in most geotechnical applications (Birmpilis et al., 2022). Like X-ray Computed tomography, Small Angle X-ray Scattering and Small Angle Neutron Scattering are techniques that have recently emerged as alternative means to study the clay microstructure. Orientation and size of the d-spacing provide in-situ measurement of bulk particle configuration on wet samples, without the need of any sample preparation (Birmpilis et al., 2019). However, it does not provide any information in terms of the spatial distribution of the fabric.

4. Experimental Strategies

To obviate the limits of existing technologies in providing a full description of the micromechanics of clays, different experimental strategies have been adopted.

Pore size distribution has been studied across different scales, spanning nanometres to millimetres, by adopting different systems. Shen et al. (2021) recently compared the pore size distribution of a shale oil reservoir sample by using MIP, Gas adsorption and Small-Angle Neutron Scattering.

In terms of imaging (e.g. X-ray Computed Tomography), a scale-up approach can be adopted. Silt grains can be used as markers when mixed into a matrix of clay, with particles too small to be visualised. Markers can be used to detect displacement within the matrix to evaluate the particle kinematic (Ibeh et al., 2021).

Imaging techniques having different resolutions, and Field of View, such XCT and SEM have been coupled in order to use a coarser analysis to understand which part of the sample was of interest, and thus worth of a finer characterisation via SEM (Desbois et al., 2017a).

To bypass the limit of destructive analyses and the difficulties in carrying out in-situ experiments, the "*sister approach*", initially proposed by Morgenstern and Tchalenko (1967), is often used. A series of identical samples was progressively sheared in a direct shear under drained condition. Each sample was stopped at different strain to allow for a full description of the evolution of the shear-induced fabric.

5. Conclusions

In the paper a short review of the challenges that the geotechnical community is facing when investigating the micromechanisms in clayey soil was presented. Alternative experimental strategies to tackle a full description of the evolution of the microstructure were proposed and described.

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