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Clay micromechanics: mapping the future of particle-scale modelling of clay

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ABSTRACT

Geotechnical engineers need to predict the macroscopic behaviour of clays in terms of strength, stiffness, and permeability and the variation in these properties during deformation. Engineers also need to predict the influence of environmental variables (temperature, pressure, pore-fluid chemistry) on these engineering properties. Particle-based methods, which explicitly model individual clay platelets and their interactions, can help identify the fundamental mechanisms that govern the engineering behaviour. Virtual samples can be created, and simulations can consider application of mechanical loading or change in the environmental conditions to generate data on particle kinematics and interparticle interactions. Particle-based models can be used to simulate stress and strain paths that are not easy to reproduce in physical experiments. This approach to simulation also enables parametric studies to understand the sensitivity of the overall behaviour to various particle characteristics and the nature of the particle interactions. This contribution provides a review of the state-of-the-art of existing particle-based models for clays, namely Discrete Element Method (DEM), Monte Carlo method (MC), and Molecular Dynamics (MD). The technical challenges, advantages, and disadvantages of each method for the simulation of clays are presented and discussed, together with the technical developments we would like to see over the next decade to realize the full potential of these modelling tools.

Keywords: clay modelling; clay micromechanics; DEM; MD; MC.

1. Introduction

Geotechnical Engineering applications rely on the use of constitutive models to reproduce and predict the stress-strain behaviour of soils at the macroscopic scale. These models are formulated under the assumption that soils are continuous media. The available engineering database of macroscopic evidence under a broad range of loading conditions allows such continuum models to capture soil responses in a variety of realistic scenarios, despite ignoring the discrete nature of soils.

In clays, changes in the environmental conditions, including pore-fluid chemistry and temperature, strongly affect the particle-to-particle interactions and particle kinematics. These fundamental processes, which are poorly understood due to a lack of particle-scale experimental evidence, significantly affect the overall or macroscopic material behaviour. Developing frameworks to investigate these mechanisms with a view to informing the advancement of constitutive frameworks is a key challenge in modern soil mechanics.

One option that is gaining traction in the geomechanics community is to use particle-based numerical methods that can model individual clay particles and interparticle interactions to simulate the behaviour of virtual systems of multiple particles. Such models can provide data on particle kinematics and interparticle interactions when the virtual samples are subjected to deformations, mechanical loading, or when there is a change in the environmental conditions. In addition, particle-based numerical models can be used to explore the behaviour upon stress and strain paths that are not easy to reproduce in physical experiments, thus supplementing existing experimental data. Parametric studies to understand the sensitivity of the overall response to key fundamental properties are also possible.

A particle-based model requires (i) a numerical framework; (ii) a description of the particle geometry; (iii) a functional form for the interaction model; (iv) data to calibrate the interaction model; and (v) a framework to validate the model. This contribution explores recent developments in particle-based modelling for clays considering these aspects of model development. The main particle-based numerical frameworks are firstly described, namely the Discrete Element Method (DEM), the Monte Carlo (MC) method, and Molecular Dynamics (MD). Then, each method and its specific use within the context of clay micromechanics is discussed. The technical challenges, advantages and disadvantages are outlined, and a state-of-the art review of the existing applications of these methods to clay modelling is presented. Finally, the technical developments necessary to address the current challenges in particle-based clay modelling, and realize the full potential of these numerical tools, are briefly presented and discussed.

2. Overview of discrete numerical methods

2.1. Discrete Element Method

The original DEM algorithm for geomechanics applications was proposed by Cundall and Strack (1979), following an earlier algorithm developed for rock mechanics applications. Cundall & Hart (1993) defined the DEM as a dynamic numerical method in which finite displacements and rotations of discrete bodies are simulated. Two types of DEM are used in geomechanics, namely block DEM (deformable bodies) and particulate DEM (rigid bodies). Of the two, particulate DEM is more prevalent in fundamental soil mechanics studies, as the assumption of rigid particles reduces the degrees of freedom and simplifies the algorithm, so that large systems of particles can be studied.

DEM numerically solves Newton's equations of motion (2nd and 3rd Newton's laws), following the principle of conservation of energy and momentum. Translational and rotational equilibrium equations are solved using an explicit, conditionally stable time integration approach. In each time increment, the contact forces acting on each particle are determined. Then, the dynamic equilibrium of each particle is considered to calculate its acceleration, and the accelerations are numerically integrated to obtain velocities and displacements. The evolution of the system is simulated by advancing forward using small time steps.

Contact forces in DEM are considered to have a normal and tangential component, determined numerically using normal and shear springs respectively, the springs can be linear or non-linear (Thornton and Randall 1988; Thornton and Yin 1991). In the contact normal direction, a small amount of overlap between particles is permitted, conceptually equivalent to the deformation occurring between real contacting bodies (contact normal deflection). The contact normal force is determined as the product of the overlapping length and the normal stiffness. A slider is included so that contact can be broken when a threshold force is reached; most often, tensile forces are not allowed unless an active reversible or irreversible adhesion force is needed. In the tangential direction, the force is calculated cumulatively as the sum of the products of the incremental (relative) displacement and the tangential spring stiffness from the initiation of the contact, thus requiring information about contacts (as well as particles) to be stored during the calculations (contact shear deflection). An upper limit on the tangential force is applied using Coulomb friction; again implemented via a numerical slider. It is worth noting that the cumulative tangential displacements in DEM simulations are non-physical as it is assumed that tangential elastic energy is conserved during rolling and before sliding occurs. Yet, the numerical implementation of such frictional laws allows reproducing key aspects of soil behaviour (Cundall 2001).

Some variations of DEM that differ significantly from Cundall and Strack's algorithm have also been used, including the contact dynamics method (Moreau 1994; Radjai and Richefeu 2009) and the implicit DEM code (Holtzman et al. 2009). Extensions to include complex particle geometry include LS-DEM (Karapiperis et al. 2020).

2.2. Monte Carlo (MC) method and Molecular Dynamics (MD)

In computational chemistry, two techniques are used to simulate the structure and trajectories of an assembly of molecules and ions, namely the Monte Carlo method (MC) and Molecular Dynamics (MD) (Alder and Wainwright 1959; Allen and Tildesley 2017). In both methods, molecules and ions are modelled as discrete (usually point-like) particles, and their interactions are described by two- or three- body potential energy functions (e. g. Lennard-Jones, Morse, Yukawa, Coulomb), describing the variation in the potential energy of the interaction with the separation of the particle centroids.

The difference between DEM force-separation laws and MC/MD potential energy functions is really an issue of semantics, as the force can be easily determined from the derivative of the energy-separation relationship. In MD, the forces derived from the potentials are used to integrate Newton's equations of motion, to derive accelerations and displacements. Conversely, in MC, particles are moved randomly to represent a target probability distribution, that is consistent with the desired state of the system. In each computational step, a randomly selected molecule (or ion, or particle) is moved to a randomly selected position, where it remains if the total energy of the system is decreased by the move. This process is repeated until the energy of the system and other calculated properties remain stable. The MC approach is not deterministic (i. e. the system does not evolve following a physically realistic path, and real particle trajectories are not generated), and can only be used to study systems in thermodynamic equilibrium.

MC and MD analysts explicitly run their simulations in ways that are consistent with probability distributions, referred to as "thermodynamic ensembles" (Ercolessi 1997). A three-letter sequence, e. g. NVE, NVT and NPT, is used to identify the ensemble used in a simulation, where N denotes the number of particles, E denotes the total energy (kinetic + potential), and V, T and P denote volume, temperature, and pressure, respectively. The quantities indicated by the letters V, E, P and T are controlled target values (Allen and Tildesley 2017). For example, in a NVE simulation, the energy is determined from the initial positions and velocities of N particles, and it is maintained constant throughout the simulation, while the volume also remains constant. In a NVT simulation, the number of particles and volume are constant, and a computational device known as a "thermostat" ensures the temperature of the system is constant. Similarly, in a NPT simulation, a "barostat" is used to control the pressure. Note that these thermodynamic ensembles are implicitly used in DEM: most geomechanics DEM simulations are in the NPT ensemble, where the temperature is set to zero, and P is replaced by a stress tensor with explicitly specified fixed and variable components.

In geomechanics applications using MD, a "coarse-grained" approach has been adopted, where the behaviour of a large number of atoms is coalesced into single units to reduce the computational cost. In contrast to DEM, most MD codes calculate the interaction energy at each time step from the separation distance alone, with no need to store information about the interactions. Furthermore, friction between particles is rarely implemented in coarse-grained MD, as it emerges from the adopted dissipation scheme and coarse-graining procedure. In MD/MC, the energy dissipation occurs collectively in the system via the barostat and/or thermostat coupling parameters, whereas in DEM the dissipation occurs locally at the contacts via friction forces. Transitioning from one formulation to the other is not trivial. This can lead to non-physical stress states as shown by Bandera (2022), where an isotropic stress state developed under 1D vertical compression when friction was neglected.

3. Application of discrete numerical methods to clay modelling

3.1. DEM clay modelling

3.1.1. Particle size and geometry

In contrast to the case of granular materials, clay particles are small (nano- to micro-metric) and elongated, with aspect ratios (length/thickness) typically larger than 10. Typical examples are clay platelets such as kaolinites, and rod-shaped nanotubes, such as halloysites. These particle geometries are responsible for some macroscopic key behaviours of clays, and therefore cannot be overlooked in discrete approaches, such as the DEM.

A key technical challenge to adopting DEM to simulate nano/micro-metric particles is related to the computational time. The small particle mass (m) and large spring stiffness (k) lead to extremely small time steps (proportional to $\sqrt{(m/k)}$ in explicit numerical integration algorithms). Some techniques have been proposed to overcome this issue, such as the multi-step method (Anandarajah 1999), or the upscaling of the particle mass (Thornton 2000). These perform well in the quasi-static regime but may pose issues for fully dynamic simulations. A potential alternative to be further explored is the use of implicit numerical integration algorithms (Jean 1999; Holtzman et al. 2009); however, convergence of implicit DEM algorithms is non-trivial and can require computationally expensive iterations (O'Sullivan, 2002).

The second challenge to using DEM in particle-based simulation of clays relates to the particle shape. The implementation of non-spherical DEM units is rather cumbersome, and the calculation of the interaction forces and moments at each contact point is nontrivial. The Euler equation for non-spherical particles is non-linear, and, Euler angles pose singularities. These limitations can be overcome by writing the rotational equation of motion in terms of quaternion dynamics (Wang and Alonso-Marroquin 2009). The implementation of elongated particles with non-circular cross sections requires the model to account for both face-to-face and edge-to-face contacts and/or interactions, which can no longer be described by an overlap and a single normal spring, but may require multiple independent variables to fully describe relative displacements (e. g. Boton et al. 2013; Boton et al. 2014). Also, highly elongated particles are prone to deform elastically (particle bending). This can be simulated by introducing internal degrees of freedom within the particles, each particle being composed of smaller units that interact via a harmonic potential. A computationally efficient alternative is to integrate the rigid body dynamics (Goldstein 1983) with the formulation of plate/beam bending (Timoshenko and Woinowsky-Krieger 1959).

Different methods have been proposed over the years to model the non-spherical nature of clay particles in DEM. The main methods, schematically showed in Fig.1, can be listed as follows:

1. Polygons and polyhedrons: The first approaches to simulate clay using DEM, which date back to the early 2000s, considered polygons or polyhedrons as the DEM unit; and this approach continues to be adopted. In two-dimensional analyses, single clay particles were simulated as assemblies of multiple rectangular elements (Anandarajah 2000; Anandarajah and Amarasinghe 2012) or segments (Khabazian et al. 2018; Bayesteh and Hoseini 2021), bonded together to behave as single units (multi-element approach, to allow for particle bending). In three-dimensions, cuboids (Yao and Anandarajah 2003) or planar-shaped irregular polyhedrons (Luo et al. 2021) were used. In most cases, the interactions are calculated in terms of a hypothetical mid plane ('common plane' technique, first proposed by Cundall in 1988). The main drawback of this approach lies in the presence of a third entity (the hypothetical mid plane) in the interaction between two particles, as an equation to calculate the potential energy associated to the mid plane interaction has yet to be proposed. In addition, polygons or polyhedrons i) cannot be implemented in most conventional DEM software, thus requiring the development of bespoke codes, and ii) they result in computationally expensive contact detection algorithms, and complex methodologies to account for relative particle orientations.

2. Clump-of-spheres particles: non-spherical shapes can be created by considering clumps of touching or overlapping circles or spheres, that behave as a single unit. This approach was adopted both in two dimensions (e. g. rod-shaped particles, Pagano et al. 2020) and three dimensions (e. g. cuboid-like particles, Jaradat and Abdelaziz (2019); hexagon-like particles, de Bono and McDowell (2022)). The fundamental advantage is that this approach inherits the benefits of circular/spherical particles, i. e. i) energy balance is always guaranteed, provided the selected timestep does not exceed the critical value for numerical stability; ii) conventional, computationally robust contact detection algorithms can be adopted; and iii) clumped-spheres particles can be easily implemented in most existing DEM codes. Importantly, this approach allows assignment of different properties to the spheres belonging to the edges and faces of each particle, allowing to account for surface non-homogeneities typical of clay particles (e. g. surface charge). While the high number of elementary units

(circles or spheres) in the simulation domain results in a high computational burden, this can be minimised by using appropriate neighbour grids, so that only relevant interactions are updated in each time step. The net interparticle interactions are obtained as the sum of all the interactions experienced by the elementary units belonging to the same particle.

3. Spheropolytopes. These are the Minkowski sum of a "polytope" (open or closed polygon/polyhedron) and a sphere (Pournin and Liebling 2005). Several authors have adopted this method to simulate granular materials made of elongated particles, e. g. sphero-segments (Azéma and Radjaï 2012), sphero-cylinders (Pournin et al. 2005), sphero-plates (Boton et al. 2013; Boton et al. 2014), and sphero-polyhedrons (Galindo-Torres et al. 2009). An example of the use of spheropolytopes to simulate clay is given by Tang, et al. (2014), where kaolinite clay particles were modelled as sphero-polygons. The main advantage is the significant reduction in data storage capacity required for the description of particle shapes, as opposed to the clump-of-spheres approach.

3.1.2. Force-separation laws

To simulate clay using DEM, the force laws cannot be based on contact mechanics as they must reflect a variety of physico-electro-chemical processes occurring between particles not yet in contact (strongly dependent on the pore-fluid chemistry and temperature), which are comparable in magnitude with mechanical interactions. The complexity of the interparticle interactions in clays has represented a limiting factor for the implementation of DEM models for clays, yet this issue is gradually being addressed by recent breakthroughs in numerical modelling.

The functional forms used in documented DEM simulations of clay (schematically shown in Fig. 1) vary regarding the extent to which they are based on the fundamental theories that can describe the interaction of colloidal particles. However, the force-separation model is developed, selecting the model parameters is nontrivial. Owing to the small size of clay particles and the complexity of the interparticle interactions, particle-scale experimental evidence to tune contact model parameters is scarce or absent.

1. DLVO-Based Models. Amongst the models that have implemented theoretically derived force-separation laws, most contributions adopt non-linear functional forms resembling the well-known potential-vs-separation function described by the DLVO (Derjaguin-Landau-Verwey-Overbeek) theory. In the DLVO theory, particles repel each other due to their interacting diffuse double layers (i.e. the charged surface and the distributed charge in the adjacent phase), while attractive interactions are to be ascribed solely to van der Waals forces. DLVO theory includes a number of simplifying assumptions, and it is strictly only able to describe the interaction between clay particles in very limited cases (e.g. for monovalent electrolyte solutions at very low concentrations, Sposito 1989; Mitchell and Soga 2005), as any long-range electrostatic attractive interactions are not considered.

Anandarajah and co-workers (Anandarajah 2000; Yao and Anandarajah 2003) implemented their force-separation law as the sum of non-linear van der Waals attraction, double-layer repulsion, and linear mechanical repulsion. To calibrate their model parameters, the van der Waals interactions were derived theoretically following the Hamaker-de Boer approach between a plate and an infinitely long wall as a function of separation distance, particle geometry, orientation, and Hamaker constant (Anandarajah and Chen 1997). The diffuse double-layer repulsive forces were derived from an approximate semi-analytical method based on the numerical computation of the electric potential over the domain (Poisson-Boltzmann equation), as a function of separation distance, particle geometry, orientation, and pore-fluid chemistry. Since the theories to compute van der Waals and diffuse double-layer interactions are not valid at very small separation distances, a cut-off distance was introduced, beyond which these interactions were considered constant. At negative separation distances (particle overlap), a linear force-displacement relation
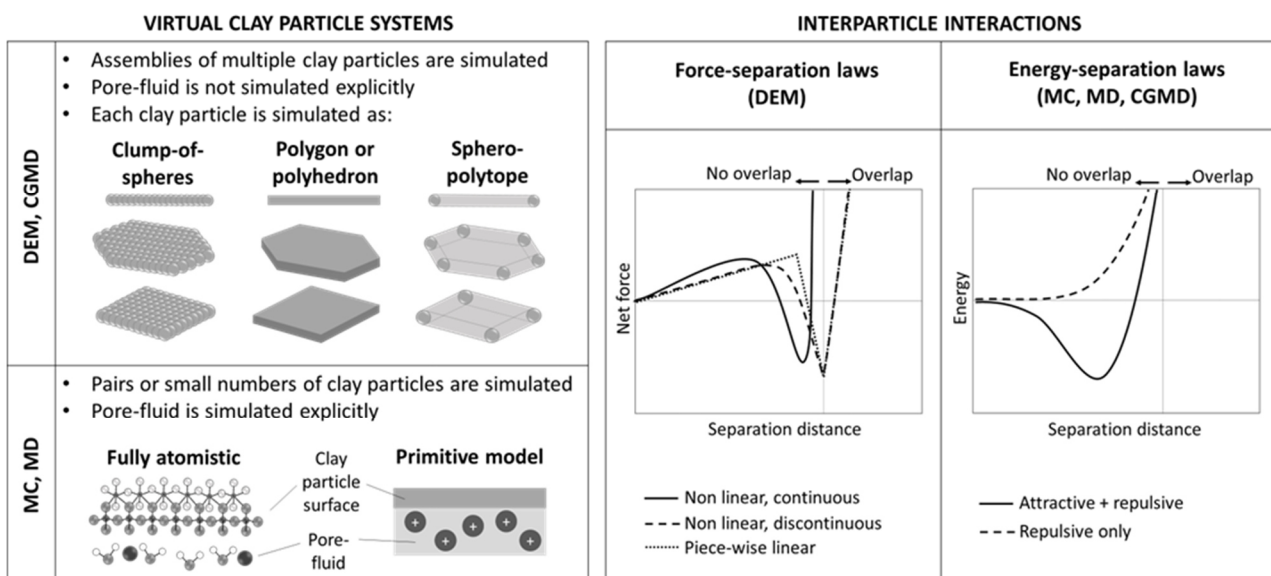

Figure 1: Examples of discrete approaches to simulate virtual clay particle systems and interparticle interactions.

was implemented to prevent interpenetration of particles. In later contributions, an additional piecewise linear law was introduced in the interlayer for the simulation of active clays (Na-montmorillonite), based on the results of MD simulations (Anandarajah and Amarasinghe 2012).

Bayesteh and co-workers (Bayesteh and Mirghasemi 2013; Khabazian et al. 2018; Bayesteh and Hoseini 2021) used Anandarajah's equations to determine the mechanical repulsion and van der Waals attraction (Anandarajah 1994; Anandarajah and Chen 1997). The repulsive double-layer forces were determined by considering Langmuir's equation between two infinite parallel particles for pure and mixed clay minerals as a function of the non-dimensional potential (accounting for pore-fluid chemistry and temperature), adjusted by applying Anandarajah's coefficients to account for particle orientations. Jaradat and Abdelaziz (2019) also implemented van der Waals attraction by following the Hamaker-de Boer approach, and double layer repulsion by considering the 3D application of the Derjaguin theory developed by Yao and Anandarajah (2003). A similar approach was adopted by Luo et al. (2021), who presented a detailed scheme to calculate the interaction (repulsive double layer and attractive van der Waals force and momentum) between platelets of irregular shapes as a function of separation distance, particle orientation, and pore-fluid chemistry.

Selecting the model parameters in DLVO-based models is non-trivial. For the van der Waals attractive force, the Hamaker constant is typically derived from the Lifshitz theory, as a function of the type of clay mineral and the pore-fluid chemistry. To determine the double-layer repulsion, some assumptions must be made, e. g. regarding the cation exchange capacity (CEC) and specific surface of clay minerals, to compute the charge density, particle thickness, surface potential and double-layer thickness, appearing in the equations to derive the repulsive force. In the absence of experimental measurements, these parameters cannot be directly calibrated, and are generally selected to fit experimental observations at the macroscale (e. g. 1D compression, Anandarajah 2000; Jaradat and Abdelaziz 2019). The remaining parameters (e. g. cut-off distances and spring stiffness at the contact) are usually selected based purely on considerations on computational costs.

2. Simple Interaction Models. As noted above, DLVO theory is not universally applicable. It is also difficult to quantify some of the parameters needed to derive the electrostatic forces, which depend on the surface charge/surface potential. These considerations, along with the difficulty of working with the analytical expressions for arbitrary particle geometries, have motivated the use of simpler force-separation models for use in DEM.

Pagano et al. (2020) proposed a simplified interaction model with piecewise linear force-separation laws. Non-contact interactions were modelled as electrostatic repulsive or attractive forces due to the presence of permanent and pore-fluid dependent charges on the surface of clay particles, and were assumed to be the only electro-chemical forces (i. e. van der Waals attraction was not considered). Adopting a multi-sphere particle geometry allowed assigning different charges to each

sphere, thus enabling the model to account for the attraction between oppositely charged edges and faces, which was not considered in previous contributions.

A similar feature was implemented by de Bono and McDowell (2022). Here, non-linear sphere-to-sphere interaction laws were designed to include van der Waals-like attraction, and long-range electrostatic repulsion or attraction, to capture different interaction regimes between spheres belonging to different clay platelets (face-to-face, edge-to-face, and edge-to-edge). As opposed to previous contributions, the functional form adopted by de Bono and Mc Dowell is continuous, and it does not allow for physical overlap (i. e. mechanical interactions are implemented as close-range repulsive forces, at positive separation distances).

For these simple force-separation laws, the model parameters do not directly represent particle or pore-fluid properties, rather they account for the effect of these properties on the evolution of the interactions in a qualitative or semi-quantitative way. Often, these parameters are selected somewhat arbitrarily, with no calibration or fitting against experimental data. One example of 'indirect' calibration of force-separation law parameters, not purely based on fitting macroscopic experimental results, appears in Pagano et al. (2020). Here, the DEM framework was designed based on a conceptual micromechanical model informed by indirect microscale experimental evidence, i. e. Mercury Intrusion Porosimetry (MIP), and Scanning Electron Microscopy (SEM) (Pedrotti and Tarantino 2017). The model parameter accounting for varying pore-fluid chemistry, i. e. the spring stiffness of the long-range electrostatic interaction, was calibrated by fitting macroscopic experimental data (void ratio at quasi-zero vertical stress) for de-ionised water as the pore-fluid. By scaling this value by the dielectric permittivity of different pore-fluids, the void ratios and relative compressibility indexes were successfully reproduced for samples saturated with different pore-fluids, rather than simply fitted.

3.2. MC and MD simulations for clay modelling

3.2.1. Atomistic simulations

At the atomic and molecular scale, two approaches can be used to determine the interactions between clay particles within a solvent (Fig. 1): (i) fully atomistic description of clay particles and electrolyte (e.g. Ebrahimi 2014), and (ii) primitive models, where only the surfaces of two clay particles are modelled as electrically charged planar surfaces, and the electrolyte is modelled explicitly in terms of ion size and charge, embedded in a uniform dielectric medium or with explicit water molecules (Carrier 2014; Dragulet et al. 2022). These atomistic approaches are computationally expensive and, hence, unsuitable to simulate systems of multiple clay particles. However, in the context of clay discrete modelling, they provide an alternative approach to the DLVO theory to generate more appropriate force-separation relationships at the atomic scale (Ebrahimi 2014; Zhu et al. 2019; Dragulet et al. 2022). These relations derive from the change of free energy as a function of the distance between two clay particles

(referred to as Potential of Mean Force, PMF, including the effect of the electrolyte), leading to an effective interparticle potential (Roux 1995).

Most documented molecular simulations of clay-water systems have focused on the crystalline swelling behaviour of the clay and on the interlayer distribution of the water and ion molecules. Cygan et al. (2004) proposed the CLAYFF force field, a general set of parameters for interaction potentials of hydrated crystalline compounds embedded in electrolytes, used in both MD and MC simulations (Ebrahimi 2014). In CLAYFF, the clay lattice is not considered to be rigid, and the total potential energy of the system has contributions from both "bonded" and "non-bonded" interactions. Bonded interactions include bond stretching and angle bending, associated with water molecules, hydroxyls, and dissolved ions. Non-bonded interactions include long-range electrostatic interactions (Coulomb), and short-range attractive interactions (van de Waals). The partial atomic charges are obtained from computational quantum mechanical modelling of well-defined structures (Density-Functional Theory, DFT), and the model parameters are empirically derived from known minerals and experimental data of crystal structures. The total energy of the system is given by the sum of the aforementioned energy terms. The most challenging species are oxygen and hydroxyl, as their charges vary depending on their occurrence in water molecules, hydroxyl groups, and bridging and substitution environments. To overcome this, CLAYFF treats them as different species depending on where they are in the system. Recently, CLAYFF has been improved to treat the edges of hydroxylated clay nanoparticles (e. g. Si-O-H, Al-O-H) (Pouvreau et al. 2019).

For the atomistic simulation of water, numerous models have been proposed (an overview is given in Pathirannahalage et al. 2021). These models consider sets of force field parameters that include the bond lengths and angles, the force constants for flexible models, the atomic partial charges, and the parameters to describe the van der Waals interactions. Some examples of water models used to simulate clay-water interactions are the MCY, TIP4P and SPC/E models (e. g. Tambach et al. 2004). In CLAYFF, water is modelled using the SPC model (Berendsen et al. 1981).

To date, very few studies have used molecular-scale derived PMFs to inform coarse-grained simulations of assemblies of clay particles. Ebrahimi et al. (2014) ran MD simulations at the atomic scale to determine the free energy at discrete separation distances for Na-montmorillonite infinite planes in water. Using a similar approach, Zhu et al. (2019) presented data on the variation in free energy per unit length with separation distance for parallel imogolite nanotubes. In both examples, the computed PMFs were used to inform coarse-grained simulations of ellipsoidal particles (Ebrahimi et al. 2014) or rod-shaped particles composed of spheres (Zhu et al. 2019), to investigate the stacking of ellipsoids, volume fraction and mechanical properties under different boundary conditions. Volkova et al. (2021) considered planar platelets of kaolinite of 10 nm in size and performed fully atomistic simulations of up to 4 structural layers under different orientations.

3.2.2. Coarse-Grained Molecular Dynamics (CGMD) simulations

In a Coarse-Grained Molecular Dynamics (CGMD) simulation, one of two approaches can be used (i) the elemental particle represents or simulates a single clay particle, or (ii) a bonded cluster of spheres represents a single clay particle.

The approach of using a single particle to represent one clay platelet was adopted by Ebrahimi (2014) and Bandera et al. (2021). Ebrahimi used the Gay-Berne potential (Gay and Berne 1981) and simulated montmorillonite platelets as flat, rigid ellipsoids (this is an inherent limitation as SEM images have indicated that montmorillonite platelets are highly flexible). Many of the Gay-Berne potential parameters do not have a physical meaning, therefore Ebrahimi calibrated the model parameters using data from the atomistic simulations described above. Simulations using the CGMD model were carried out using LAMMPS (Plimpton 1995) and restricted to considering the elastic properties of an assembly of particles.

Bandera et al. (2021) applied Ebrahimi's modelling framework to kaolinite, again using LAMMPS. Arguably, this is a more appropriate target mineralogy for this modelling framework as kaolinite particles can be more realistically modelled as flat, rigid ellipsoids. Bandera et al. modified the functional form of the potential to only include the repulsive term in the Gay-Berne potential, and calibrated their data against the DLVO theory. Key issues raised in Bandera et al. (2021) and Bandera (2022) include the need to simulate large systems ($\approx$ 100,000 particles) to capture the meso-structures that develop in clay, and the ambiguity about how to simulate or account for interparticle friction.

Liu et al. (2015) used rigid clusters comprising 122 spheres arranged in a hexagonal geometry to simulate suspensions of kaolinite particles, implemented in LAMMPS. The interaction parameters were informed by the DLVO theory. Sjoblom (2016) and Amipour and Sjoblom (2019) also used rigid clusters of spherical particles. Again, the DLVO theory was used to inform the interaction model adopted, and linear contact springs were included to simulate Born and Stern-layer repulsion at short interaction distances. One-dimensional compression was considered in Sjoblom (2016), while Aminpour and Sjoblom (2019) considered triaxial compression.

4. Conclusions and perspectives

While clays are traditionally modeled in terms of their constitutive behavior (Mitchell and Soga, 2005), discrete modeling is essential for a micromechanical approach. Discrete numerical methods, namely DEM and coarse-grained MD, provide a promising framework. These methods, originated from different technical perspectives, are algorithmically similar, as they consider the dynamics of a system of discrete bodies at discrete time intervals, and use an explicit time integrator to simulate the evolution of the system. The key differences are: (1) DEM uses force-separation laws, and the history of the interaction is recorded. In MD, interactions are modelled as the variation of the potential energy with

respect to the particle centre-centre separation, with no need to store information about the interaction history. (2) In DEM, the frictional contacts between particles are modelled by means of shear springs, with frictional and visco-elastic dissipation at the contact points. In MD, simulations consider the use of ensembles, and only bulk dissipation is considered; the geometry and steric exclusions are included in the interactions.

Presently, several key issues remain around the particle-scale simulation of clays:

1. Computational cost: the small time step and need of large numbers of particles to capture the formation of clusters/aggregates are challenging aspects. Single-platelet approaches with one interaction calculation per contacting pair are advantageous. DEM or MD software parallelized with MPI to access distributed memory on high performance computers, or multiple cores on individual workstations, can readily access the hardware that is now available. GPUs may also provide a solution.

2. Particle shape and flexibility: particle shape as a rigid or flexible unit is crucial. To account for clustering and pressure-dependence, the anisometric (elongated) shape may be sufficient, and should be appropriately defined for the material to be simulated. When relevant, particle flexibility should be accounted for by introducing internal degrees of freedom.

3. Uncertainty around the fundamental interactions: two key areas for development are (i) use of atomic force microscopy to obtain experimental data on surface charge/surface potential, and (ii) additional atomistic MC/MD simulations for a range of mineralogies, pH values, and electrolyte concentrations.

4. Time-dependent behaviour: thermal energy may be negligible in clays compared to cohesion energy, and therefore irrelevant for the mechanical behaviour. Still, it can play a significant role in long-time phenomena (creep, relaxation), which may be modelled by means of thermally-activated interactions (Kwok and Bolton 2010)

5. Calibration: input parameters in discrete simulations are defined at the particle scale, therefore a direct calibration against experimental data at the particle scale is required. This will not be possible until a significant breakthrough in particle-scale experimental techniques for clays is achieved. For a breakthrough in clay discrete modelling, benchmark tests need to be defined, e. g. the size of the representative elementary volume (REV) to reproduce the stress-strain behaviour with non-spherical particle shapes and colloidal interactions must be determined. The comparison of several key features (effect of pre-consolidation pressure, visco-plastic response, evolution of void ratio) obtained from simulations and experiments can provide a means to calibrate the interactions.

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