

Chemo-mechanical coupling in fine-grained soils and sedimentary rocks

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Introduction

A recurrent theme in the use of the subsurface for water, energy, or carbon mitigation is the importance of fine-grained (clay-rich) soils and sedimentary rocks. Fine-grained rocks (shale, mudstone) are used as caprocks, host rocks, and source rocks in a range of low-carbon energy technologies including carbon capture and storage, high-level radioactive waste storage, and shale hydrocarbon extraction. Fine-grained soils play an equally out-sized role in agriculture and soil carbon storage. This importance of clay-rich media derives largely from their distinct hydrologic and mechanical properties (ultra-low permeability, swelling-shrinking and cracking). These properties are of singular importance in controlling fluid flow in the subsurface (where fine-grained rocks constitute roughly two-thirds of the sedimentary rock mass), yet they remain a sparsely charted frontier research area in groundwater hydrology.

Here, we review existing data on the hydrologic and mechanical properties of fine grained soils and sedimentary rocks. We show that these properties are controlled by chemo-mechanical-chemical couplings at two key scales: a nanoscale defined by the aggregation and swelling of clay (nano)particles and a mesoscale defined by the assemblage of larger grains (Bourg and Ajo-Franklin, 2017). We present recent advances in modeling the distinct properties of clay-rich media on these two length scales using molecular dynamics (MD) and computational fluid dynamics (CFD) simulations

Macroscale view

Experimental data on the core-scale properties of fine grained soils and sedimentary rocks reveal two important features. First, these media display a well-known sensitivity of their mechanical and hydrologic properties to pore water chemistry. This sensitivity reflects the influence of clay swelling, a phenomenon that occurs over length scales of tens of nanometers as dissolved ions screen the electrostatic repulsion between negatively charged clay nanoparticles.

Second, these media display a significant threshold in their hydrologic and mechanical properties at a clay mineral mass fraction of $\sim 1/3$ (Fig. 1). This threshold is consistent with a simple conceptual model of the structure of sediments and sedimentary rocks on length scales of tens of micrometers on which these media can be viewed as a mixture of large grains (quartz, feldspar, carbonate), macropores, and a fine-grained nanoporous clay matrix (Bourg, 2015).

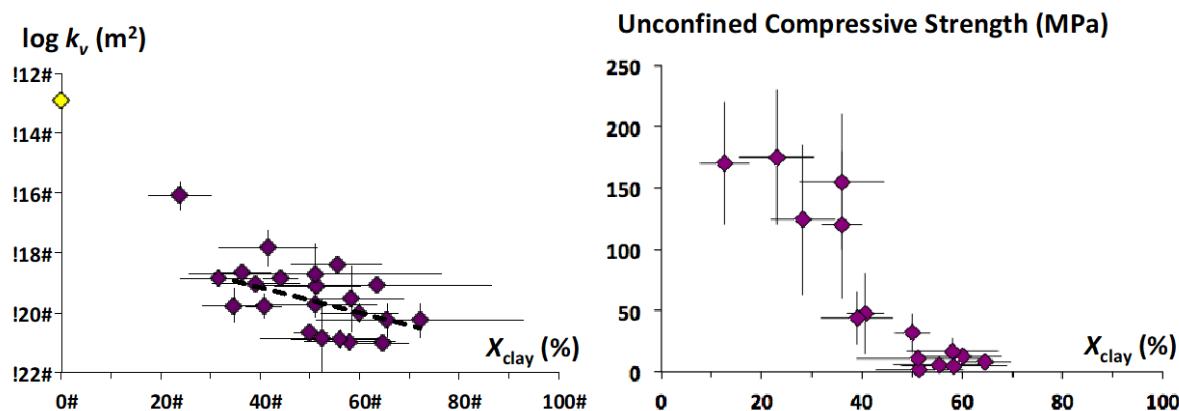


Fig. 1: Compilation of core-scale experimental data on the permeability and unconfined compressive strength of shale and mudstone as a function of clay mineral mass fraction.

A key challenge in describing the macroscale properties of fine-grained soils, sediments, and sedimentary rocks, then, is the fact that these properties reflect coupled chemical, hydrologic, and mechanical properties at two key length scales: 1) a mesoscale (micrometers to tens of micrometers) characteristic of the distribution of macropores and microporous clay within an assemblage of coarse grains and 2) a nanoscale (nanometers to tens of nanometers) characteristic of the internal properties (mechanics, permeability) of the microporous clay matrix.

Mesoscale view

Development of a mesoscale model of fine-grained soils and sedimentary rocks requires a modeling framework capable of capturing 1) the co-existence of coarse grains, macropores, and microporous clay and 2) the ability of the clay matrix to swell, shrink, and deform in response to fluid chemistry and hydrology. Our approach relies on computational fluid dynamics (CFD) simulations to describe these phenomena. More precisely, we couple the Darcy-Brinkman equation with constitutive relations describing the stress-porosity, porosity-permeability, and porosity-rheology relations of the microporous clay matrix as a function of pore water chemistry. Preliminary results suggest that our modeling approach captures several important features of fluid flow in fine-grained sedimentary rocks.

Nanoscale view

As noted above, a key set of inputs of our meso-scale model are the constitutive relations describing the stress-porosity, porosity-permeability, and porosity-rheology relations of the microporous clay matrix as a function of pore water chemistry. Our approach relies on molecular dynamics (MD) simulations to predict these constitutive relations. Our first MD simulation approach uses the metadynamics methodology to determine the free energy of swelling of a pair of smectite clay nanoparticles in liquid water as a function of aqueous chemistry, which yields the relationship between swelling pressure and pore width. Our second MD simulation approach consists in modeling the progressive dehydration of large assemblages of tens of smectite clay nanoparticles (Fig. 2) in order to predict their structural and transport properties as a function of porosity and aqueous chemistry.

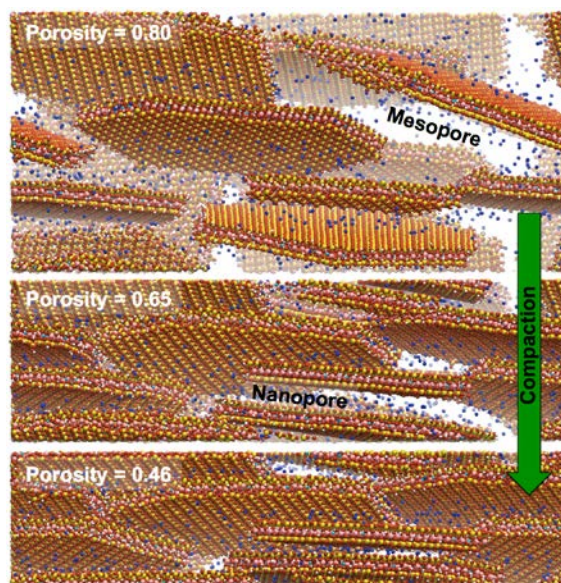


Fig. 2: MD simulation of the progressive dehydration of clay-water mixture containing ten smectite clay nanoparticles (water molecules are not shown; blue spheres are exchangeable Na ions; smectite particles are shown as yellow, red, pink, and cyan spheres).

References

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