Structure and dynamics in clays from molecular simulations

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Clays are layered alumino-silicate minerals involved in many industrial and environmental processes. In particular, their possible use as confinement barriers for the geological disposal of radioactive waste has motivated a large number of experimental and theoretical studies on the retention and transport of water and ions in clays. These properties are quantified by empirical parameters (sorption constants, effective diffusion coefficients) measured at the macroscopic scale that need to be related to the underlying microscopic mechanisms. One difficulty in understanding the latter arises from the complex, multi-porosity structure of clay materials. Most clay layers, such as Montmorillonite, bear a negative charged due to isomorphic substitutions, compensated by counterions. Stacks of parallel layers form finite particles. Counterions are located in the (interlayer) nano-porosity between particles, but also inside the larger pores between particles. Cations in these large pores can sorb onto the external (basal) surfaces of particles and also be exchanged with interlayer cations via the particle edges. We will show how molecular simulations allow to characterize on the nanoscopic scale the specific interactions between ions and clay surfaces, and to estimate the diffusion of mobile species (water and ions) inside and outside the clay particles. Moreover, transfer rates between the interlayer and the interparticle pores via the lateral surfaces can be evaluated. Our results demonstrate that, as expected, anions are excluded from the interlayer spaces, while virtually no activation is observed for the exchange of cations and water between the two porosities. This justifies the averaging procedures used to interpret macroscopic tracer diffusion data in compacted, water-saturated bentonites. Finally, the dynamical quantities obtained by molecular simulations can be included in macroscopic models accounting for the complex geometry of the material to be compared with experimental diffusion data.

Archaean-Proterozoic evolution in East Antarctica

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The Grunehogna Craton (GC), western Droning-Maud Land (East Antarctica) was part of the Archean to Palaeo-protero¬zoic Kalahari Craton of Southern Africa prior to the Jurassic breakup of Gondwana. The basement of the GC is almost entirely covered by ice and is only exposed within a small area (2x4 km^2) comprising the leucocratic two-mica Annandags¬toppane granite crosscut by garnet-bearing pegmatite dykes. Less common are darker varieties of Bt granite, Bt-rich cumulate fragments and Jurassic (?) basalt dykes. The granite (and hence the craton) has been dated previously only by Rb-Sr and Pb-Pb mica and whole-rock methods [1; 2]. Employing the novel technique of electric-pulse fragmentation (using SelFrag®), we have separated zircon from different samples of the granite. U-Pb dating of these zircons by LA-ICP-MS resulted in a crystallisation age of the granite of 3,067 ±22 Ma. Inherited grains were found to be ~200 Ma older. The crystallisation age is in agreement with the results from the previous Rb-Sr studies [1, 2] and is coeval with the granites and rhyolites of the Dominion Group (Witwatersrand) in South Africa.

The sedimentary cover of the GC is well exposed in a 250 km long mountain range at its eastern margin and consists of the Mesoproterozoic Ritscherflya Supergroup. It was deposited in a marine tidal environment at 1,100 ±30 Ma [e.g., 3], exceeds 2,500 m in total thickness and is siliciclastic with volcanic intercalations. Detrital zircons in the clastic sediments are rounded due to sedimentary transport (100–400 µm in length) and show a large variety of internal zoning patterns. Preliminary results on their age distribution show the dominant peak at 1,110 to 1,180 Ma, i.e., close to the sedimentation age and contemporenous with the arc magmatism of the Grenvillian orogeny of the adjacent metamorphic Maud belt, which bounds the GC to the East and South. Older age peaks in the Ritscherflya sediment zircons include those at ~2,040 Ma and at ~2,700 Ma. Zircons corresponding to the age of the basement granite have not been found, yet.

[1] Barton *et al*. (1987) *CMP*, **97**: 488–496. [2] Halpern (1970) *Science*, **169**: 977–978. [3] Jones *et al*. (2003) *Tectonophysics*, **375**: 247–260.