In-situ visualization of molecular self-assembly and surface restructuring at the mineral-water interface

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Organic additives at the mineral-water interface can decisively effect dynamic processes like, for example, dissolution, growth and surface restructuring. Elucidating the interplay of organic molecules with mineral interfaces and the formation of self-assembled structures on the surface in liquid environments is furthermore of utmost interest within biomineralization research [1]. Insights into the interfacial structure at the nanoscale can be obtained by using atomic force microscopy (AFM). This powerful imaging technique has proven to provide atomic and molecular-resolution images of mineral surfaces, even during *in-situ* measurements in liquid environments [2].

Here, we present high-resolution AFM studies on calcite(10.4) in aqueous solutions where we focus on the effect of the presence of organic calcium chelating agents. We discuss the entanglement of molecular self-assembly [3] and additive-induced surface restructuring [4]. Interestingly, the molecular arrangement of the additives on the calcite(10.4) surface as well as the dynamic restructuring of the surface itself may be observed at the same time, indicating that self-assembly and surface restructuring are difficult to disentangle. In contrast to the surface restructuring, which was found to be independent of the pH value in our experiments, the formation of molecular structures at the mineral-water interface was revealed to sensitively depend on the actual pH.

Our findings illustrate the complexity of interdependent dynamic processes at the calcite(10.4)-water interface in the presence of organic molecules. Therefore, the results contribute to a fundamental molecular-level understanding and disentanglement of interfacial processes and may pave the way for a precise prediction of these processes at mineral-water interfaces.

[1] Ricci *et al*. (2015) *Langmuir* **31**, 7563-7571. [2] Rode *et al*. (2009) *Langmuir* **25**, 2850-2853. [3] Schreiber *et al*. (2013) *Soft Matter* **9**, 7145-7149. [4] Momper *et al*. (2015) *Langmuir* **31**, 7283-7287.