## Interaction between barite and celestite cleavage surfaces and carbonate bearing aqueous solutions

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In nature, crystals of the witherite-strontianite solid solution series often show a significant chemical bimodality, i.e. intermediate compositions in this solid solution are very rare. Such a chemical bimodality can not be easily justified by invoking thermodynamic arguments. However, it has been suggested that witherite-strontianite crystals can result from the replacement of crystals of the barite-celestite solid solution which act as precursors. Consequently, the chemical bimodality in the carbonates might be a compositional feature inherited from the original sulphates. In this case, the replacement implies the coupling of dissolution and crystallisation reactions occurring on the interface between the  $Ba_xSr_{1-x}SO_4$  crystal surfaces and carbonate bearing aqueous solutions.

In this work, we present in situ Atomic Force Microscopy (AFM) observations of the interaction between barite and celestite (001) and (210) faces with static  $Na_2CO_3$  aqueous solutions at room temperature. The concentrations of the solutions used were in the range between 0.001 mol/l and 0.1 mol/l. Typically, such interaction is characterised by the dissolution of the original faces and the rapid nucleation and spreading of islands (~ 3 nm in height). The coalescence of the islands leads to the formation of a homogeneous epitaxial layer. Measurements on series of AFM images provide quantitative information about coupled dissolution-growth rates. On the basis of our results, we discuss the viability of the transfer of chemical information at a nanoscale during the replacement of the barite-celestite crystals by whitherite-strontianite.

## Experimentally determined biomediated Sr<sup>2+</sup> partitition coefficient for dolomite

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Aerobic culture experiments with two strains of moderately halophylic bacteria grown in solid medium have been used to determine the  $Sr^{2+}$  partition coefficient for dolomite at different temperatures and chemistries. The Sr<sup>2+</sup>, Ca<sup>2+</sup> and Mg<sup>2+</sup> data and results from XRD studies indicate that Sr<sup>2+</sup> incorporation into dolomite crystals is not by substitution of Ca2+, but instead, in our experiments, Ca2+ is replaced by Mg<sup>2+</sup>. Sr<sup>2+</sup> enrichment is probably via surface adsorption or occlusion. Thus, the experimental Sr<sup>2+</sup> partition coefficient between dolomite and fluid is represented here by the ratio of the Sr<sup>2+</sup> concentration in the solid phase (dolomite, "dol") over that in the fluid (site of dolomite precipitation = bacteriamedium interface, "bmi"),  $D_{Sr}^{dol} = Sr_{dol}/Sr_{bmi}$ . Together, our observations suggest that  $D_{Sr}^{dol}$  is apparently related to microbial activity, temperature and sulfate concentration. Therefore, the highest values calculated in the experiments, in comparison with the theoretical values, have implications for interpretation of depositional environment of ancient dolomite. The results of our cultures experiments show that high very high Sr<sup>2+</sup> concentrations in ancient dolomite could reflect microbial mediation.