



## **Nanoscale observations of coupled growth and dissolution on celestite {001} surfaces in contact with carbonate-bearing aqueous solutions**

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In this work, we present in situ Atomic Force Microscopy (AFM) observations of the interaction between celestite ( $\text{SrSO}_4$ ) {001} surfaces and static  $\text{Na}_2\text{CO}_3$  aqueous solutions. The analysis of sequences of AFM images show that the interaction is characterized by a rapid alteration (carbonatation) and dissolution of the original celestite (001) surface, shortly followed by the formation of strontianite ( $\text{SrCO}_3$ ). The crystallization of this new phase involves the formation of islands, which preferentially occurs on steps of the dissolving celestite substrate. Strontianite islands rapidly grow in height, reaching a significant thickness ( $\sim 3$  nm), which remains almost constant during their subsequent lateral spreading. AFM observations indicate that the growth of strontianite islands on celestite (001) surface has an epitaxial character. Assuming a parallelism between celestite and strontianite orthorhombic unit cells as the most probable epitaxial relationship, the following misfits have been calculated: 4% along [100] and 0.6% along [010]. These low misfits indicate a good matching between both structures through the interface. The progressive coalescence of islands leads to the formation of a quite homogeneous strontianite epitaxial layer, which partially armours the substrate from further dissolution. However, before this occurs celestite (001) dissolution kinetics is enhanced. Our measurements on series of AFM images demonstrate that at the early stages of the strontianite layer formation, dissolution rates of monosteps ( $\sim 3.4 \text{ \AA}$ ) on celestite (001) face increase up to 140% for moderate carbonate concentrations in the aqueous solutions. This clearly indicates a coupling between celestite dissolution and strontianite growth.