

Calcite surface renewal caused by fluid inclusions mobility: an AFM study.

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Diffusion of elements from the bulk to the surface of crystal has been recognized at elevated temperature and pressure but it has been always underestimated at standard conditions. In this work, we present evidences of spontaneous surface renewal due to fast ion mobility (1-2 min) originating from fluid inclusions accumulated in crystallites on the topmost layers. The evolution of {1014} freshly cleaved surface of a calcite crystal containing fluid inclusions was observed by an Atomic Force Microscope (AFM) under a weak mechanical strain (applied force < 10 nN).

Continuous *in situ* images obtained under air conditions (relative humidity $\approx 70\%$) show spontaneous nucleation and fast growth of hillocks along the cleavage plan. Hillocks germination occurs randomly on the atomically flat surface with an average height of $9 \pm 2 \text{ \AA}$ and grows preferentially along horizontal direction. After 15 minutes of AFM scanning, the growth process ends and the calcite surface is partially recovered with these hillocks with no evidences of dissolution pits.

This phenomenon is interpreted to correspond to the movement of the ions and the CO₂ degassing from the lattice to the crystal surface along grain boundaries or microfractures. We estimated the ion mobility rate was $10^{-10} \text{ m.s}^{-1}$ independently of the mechanical strain applied on the calcite sample. Although the relatively weak pressure of the AFM tip on the calcite, ions can move extremely quickly and contribute to change drastically the surface properties. The observed dynamic surface of calcite has important implications in conceptual models for the behaviour of the water/carbonate mineral interface.