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**Supporting Information**  
**For**  
**Direct Observations of the Occlusion of Soil Organic Matter within**  
**Calcite**

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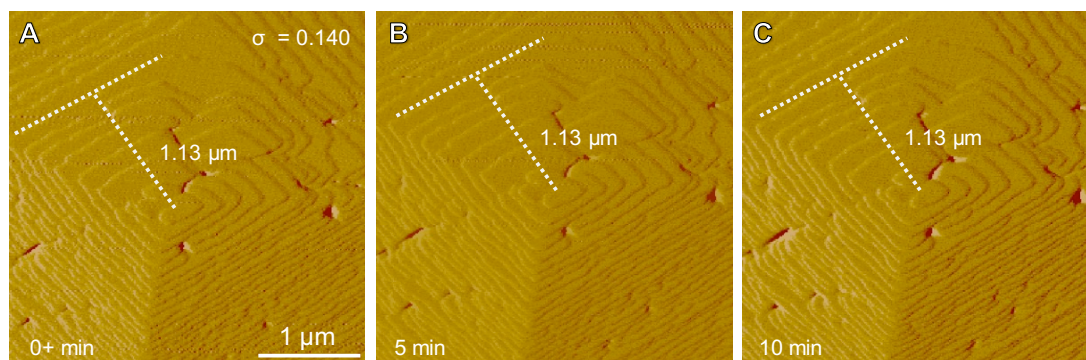
**10 pages; 2 tables; 11 figures**  
**SI tables (S2)**  
**SI figures (S2-S10)**

**Table S1.** Solution composition used in the oxalate-containing experiments.

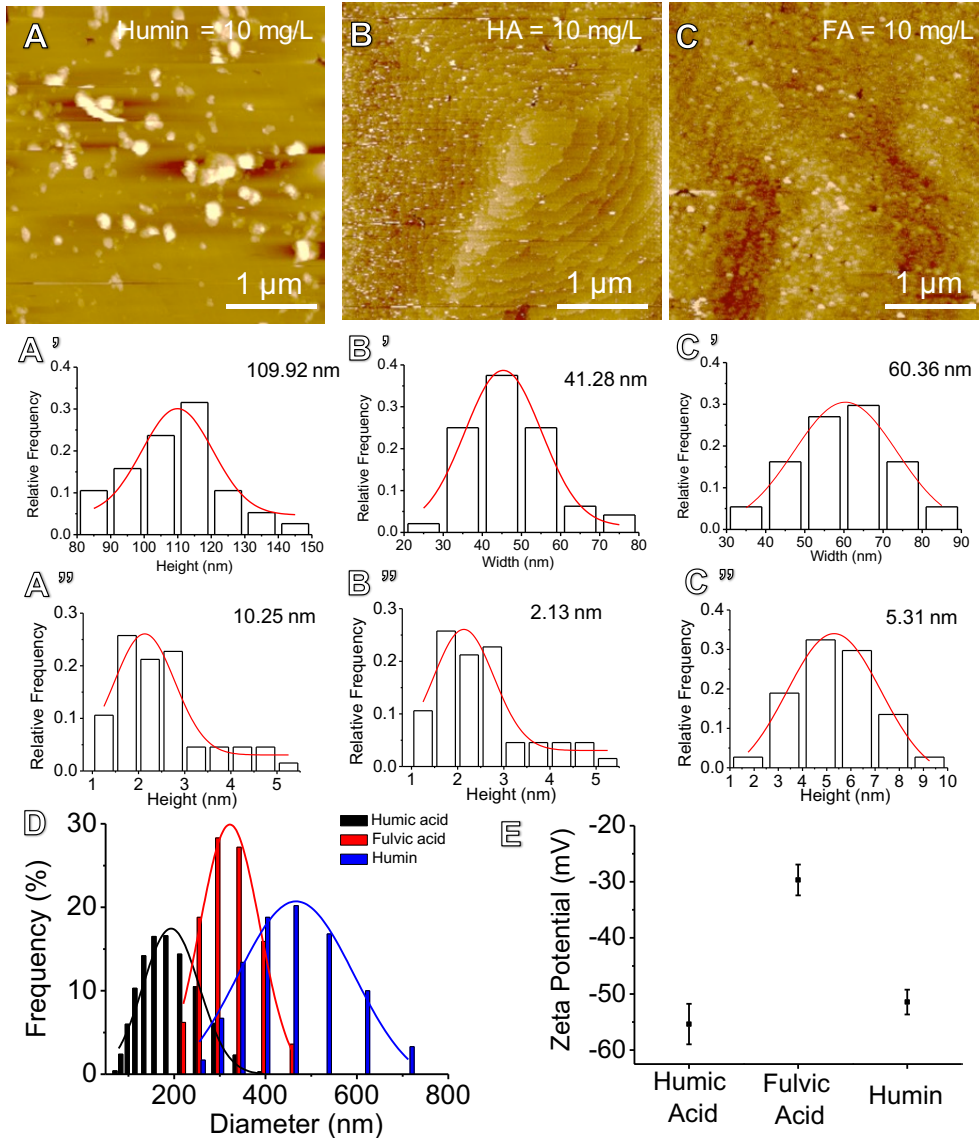
IS	pH	[Ca <sup>2+</sup> ] (mM)	[C <sub>2</sub> O <sub>4</sub> <sup>2-</sup> ] (mM)
0.11	8.3	0.17	0.1

**Table S2.** The supersaturation of two calcium oxalate phases.

phase	$\sigma$
calcium oxalate monohydrate (COM)	-0.240
calcium oxalate dihydrate (COD)	-0.484

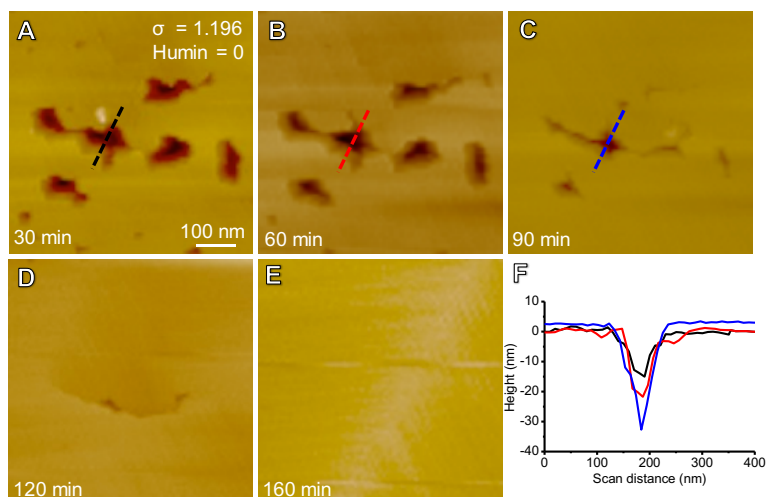


**Figure S1.** Time-resolved AFM images showing that the same step does not move in a near-equilibrium supersaturated solution ( $\sigma = 0.140$ , pH 8.3).

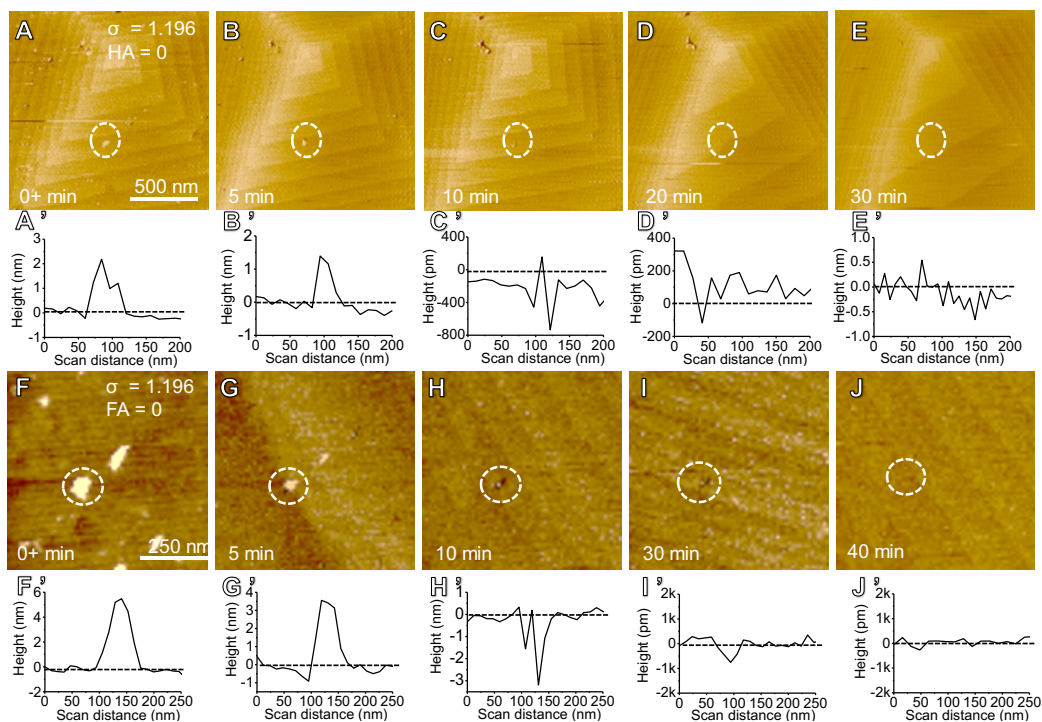


**Figure S2.** (A-C) AFM images (contact mode) after 20 min of injection of a supersaturated solution ( $\sigma=0.140$ , pH 8.3) in the presence of (A) humin, (B) humic acid (HA), or (C) fulvic acid (FA). The size distributions of the aggregated (A') humin, (B') HA, and (C') FA particles adsorbed on the calcite surface with an average width of  $109.92 \text{ nm} \pm 2.08 \text{ nm}$  ( $n = 40$ ),  $41.28 \pm 1.06 \text{ nm}$  ( $n = 40$ ) and  $60.36 \pm 0.48 \text{ nm}$  ( $n = 40$ ), respectively, and with heights of  $10.25 \pm 0.31 \text{ nm}$  ( $n = 50$ ),  $2.13 \pm 0.11 \text{ nm}$  ( $n = 40$ ) and  $5.31 \pm 0.06 \text{ nm}$  ( $n = 40$ ), respectively, measured from AFM in situ images. (D) The size distribution of three humic substances at 10 mg/L in a near-equilibrium supersaturated solution with respect to calcite ( $\sigma=0.140$ , pH 8.3) measured by DLS. The average diameters of HA, FA and humin are  $193.24 \pm 4.55 \text{ nm}$  ( $n = 4$ ),  $321.99 \pm$

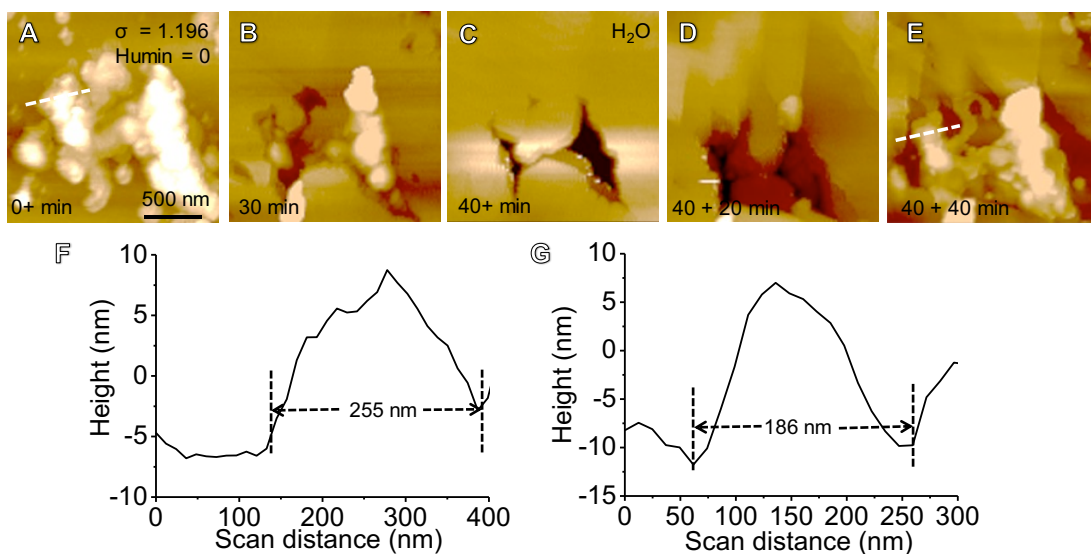
4.53 nm ( $n = 4$ ) and  $466.88 \pm 8.14$  nm ( $n = 4$ ), respectively. (E) The zeta potential of three humic substances at 10 mg/L (pH 8.3).



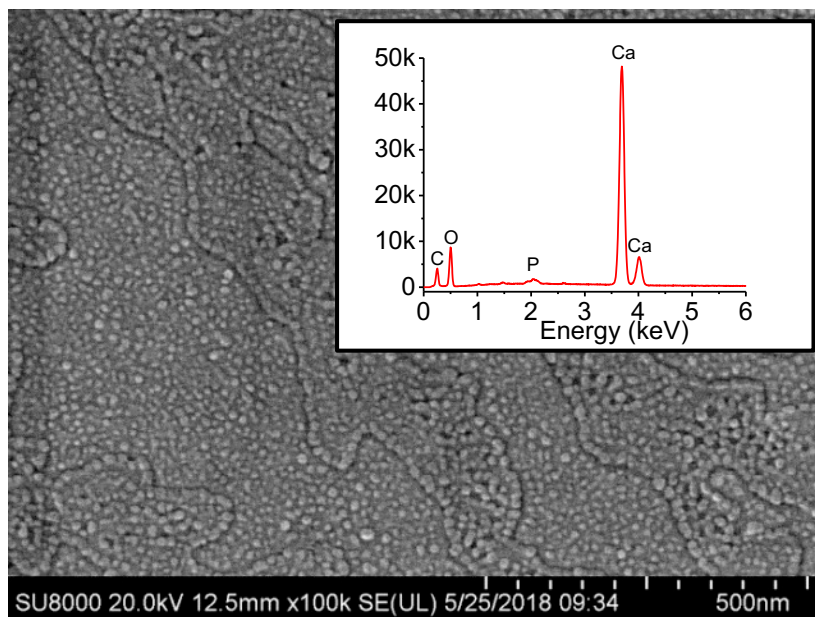
**Figure S3.** AFM images of the detailed process of the cavities closing as shown in Figure 1. (A-E) The cavities were gradually closed, indicating the presence of internal cavities. (F) The depth of the cavities measured along the dashed lines in A-C The cavities were gradually deepened from 14 nm in (A) to 36 nm in (C).



**Figure S4.** Incorporation dynamics of humic acid and fulvic acid within calcite. A time sequence of AFM height images showing a detailed process of (A-E) humic acid (HA) particles or (F-J) fulvic acid (FA) particles buried into calcite during growth by step advancement in a supersaturated solution ( $\sigma = 1.196$ , pH 8.3). (A'-J') Height profiles of humic acid or fulvic acid particles in the dotted circles in (A-J) showing corresponding height changes during the occlusion process.

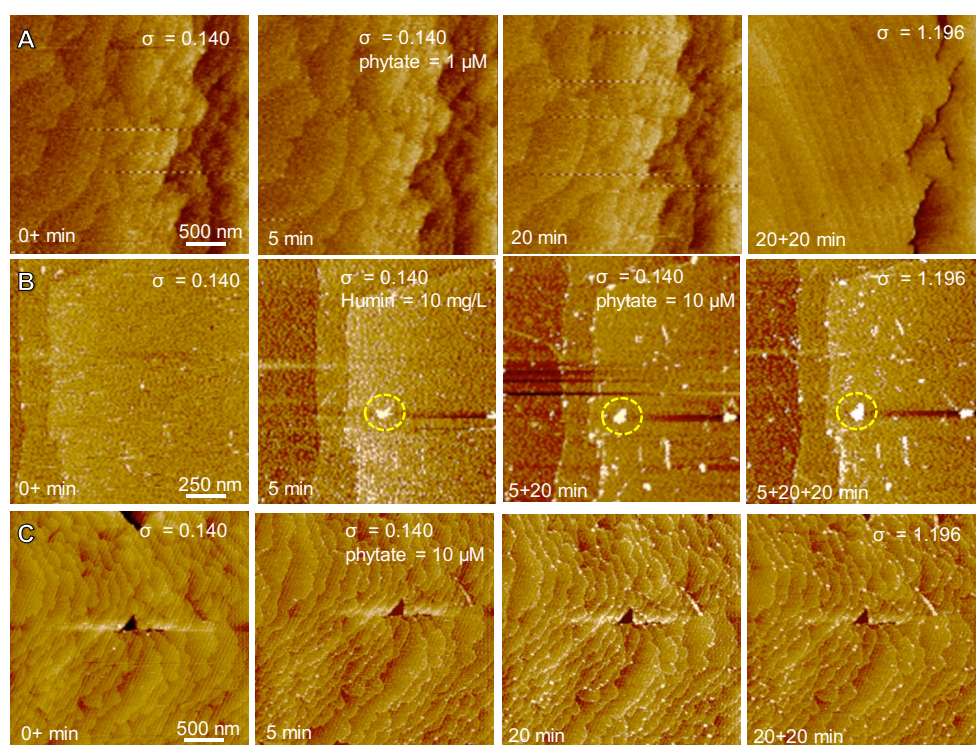


**Figure S5.** (A-C) A time sequence of AFM height images showing humin particles buried into the calcite in a supersaturated solution ( $\sigma=1.196$ , pH 8.3). (D, E) Upon introduction of ultrapure water, the same area of calcite rapidly dissolves and the buried particles are exposed. (F-G) The width of the same particles measured along the white dashed lines in (A and E) decreases after the occlusion is reexposed.



**Figure S6.** SEM image of the precipitated particles formed on the calcite surface in a solution supersaturated with respect to calcite ( $\sigma=0.140$ ) in the presence of 100  $\mu\text{M}$  phytate for 12 h. (Inset) EDX spectrum showing that the particles consist of Ca, O, C, P.

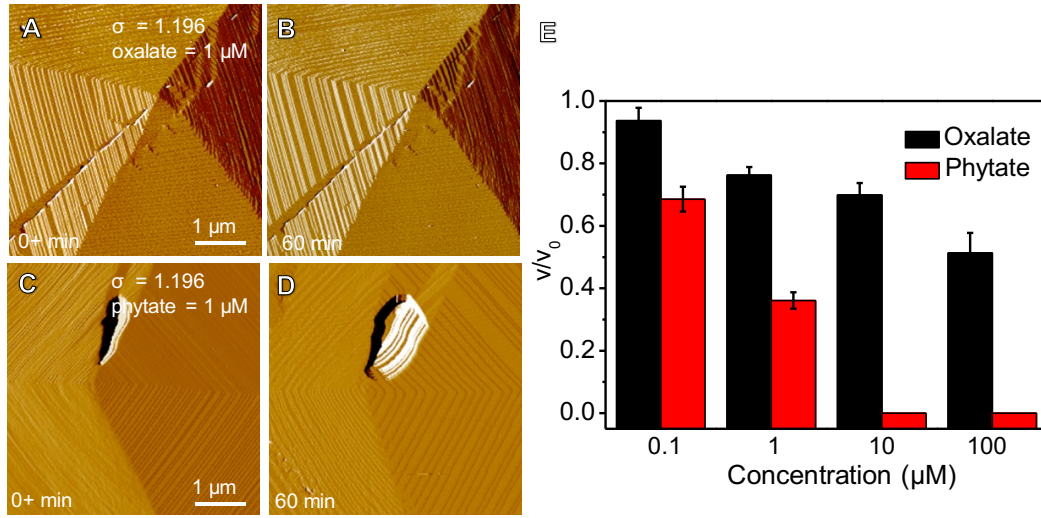




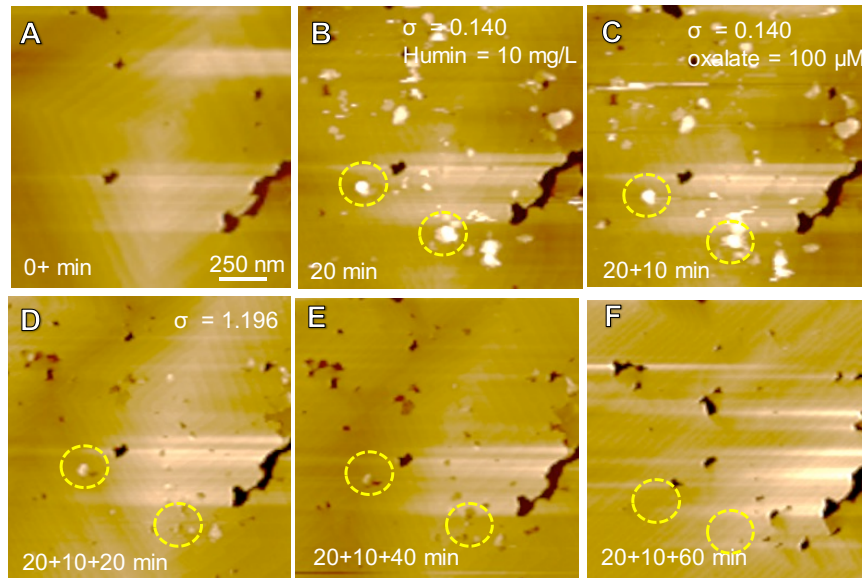
71 **Figure S7.** (A) AFM images showing that no new particles are formed in a  
 72 supersaturated solution at  $\sigma = 0.140$  in the presence of  $1 \mu\text{M}$  phytate and the steps  
 73 could advance in a highly supersaturated solution ( $\sigma = 1.196$ , pH 8.3). (B) The effect  
 74 of phytate ( $10 \mu\text{M}$ ) on the occlusion process. After the introduction of  $10 \mu\text{M}$  phytate  
 75 in a supersaturated solution ( $\sigma = 0.140$ , pH 8.3), newly formed particles are rapidly  
 76 adsorbed along the step edges to inhibit the burial of humin in high supersaturation  
 77 calcite solution ( $\sigma = 1.196$ , pH 8.3). (C) *In situ* AFM height images showing the  
 78 adsorption of particles on the step edges of a growing spiral in a supersaturated  
 79 solution ( $\sigma = 0.140$ , pH 8.3) in the presence of  $10 \mu\text{M}$  phytate; the step growth is  
 80 inhibited and cannot advance in a highly supersaturated solution ( $\sigma = 1.196$ , pH 8.3).

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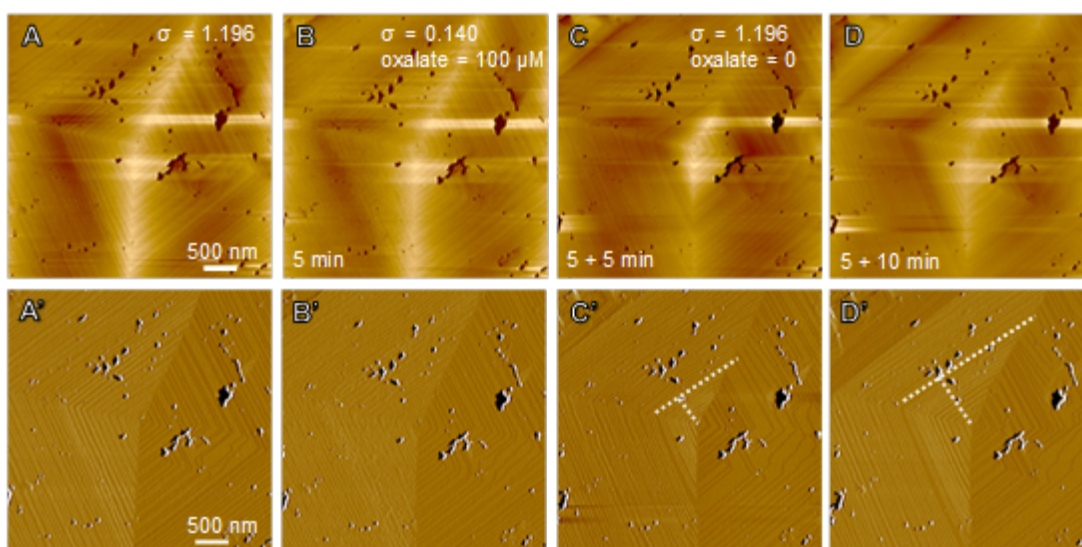
84 **Figure S8.** (A-B) AFM deflection images in contact mode of calcite hillocks grown in  
 85 the presence of 1 μM oxalate at  $\sigma = 1.196$ , pH 8.3. (C, D) AFM deflection images of a  
 86 calcite hillock in the presence of 1 μM phytate at  $\sigma = 1.196$ , pH 8.3 in contact mode.  
 87 (E) Relative step movement velocities  $v/v_0$  along  $v_+$  or  $v_-$  in the presence of different  
 88 concentrations of phytate or oxalate in a supersaturated solution ( $\sigma = 1.196$ , pH 8.3),  
 89 indicating the inhibitory effect of phytate.



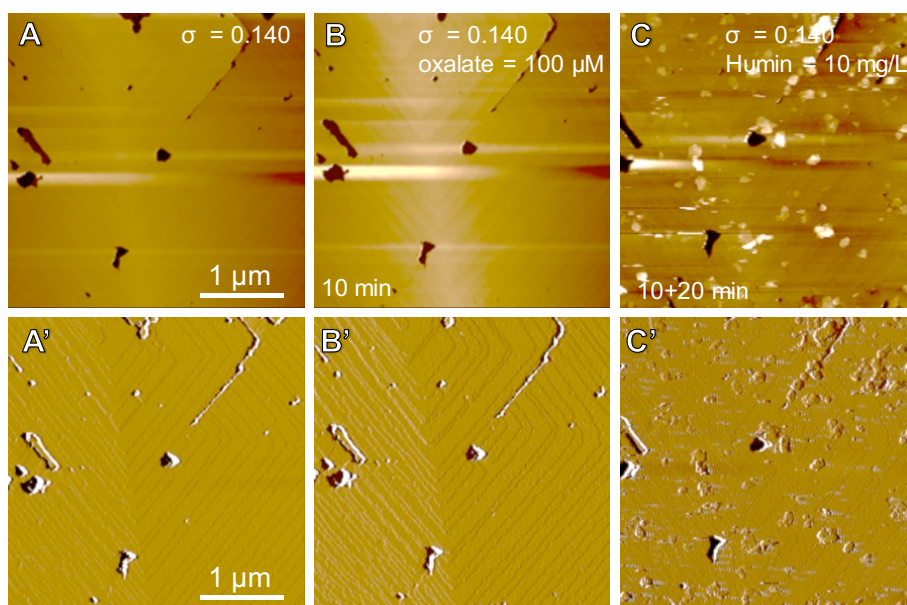
90 **Figure S9.** The effect of oxalate on the occlusion process. (A, B) AFM images of  
 91 humin adsorbed on a calcite surface to form particles (about 10 nm in height) after 20  
 92 min of exposure in a supersaturated solution ( $\sigma = 0.140$ , pH 8.3) in the presence of 10



mg/L humin. (C) No other new particles formed after 10 min of addition of 100  $\mu$ M oxalate in a supersaturated solution ( $\sigma = 0.140$ , pH 8.3). (D-F) Following 60 min of injecting a highly supersaturated solution ( $\sigma = 1.196$ , pH 8.3), the adsorbed humin particles are gradually incorporated into a calcite spiral.



**Figure S10.** AFM (A, B) height and (A', B') deflection images illustrating that no new particles are formed in a supersaturated solution at  $\sigma = 0.140$  in the presence of 100  $\mu$ M oxalate. AFM (C, D) height and (C', D') deflection images showing that the spiral growth recovers with step advancement (indicated by the white dotted lines in C' and D' after injecting a high supersaturation solution ( $\sigma = 1.196$ ) without oxalate.



**Figure S11.** AFM (A, B) height and (A', B') deflection images of a calcite surface in a supersaturated solution ( $\sigma = 0.140$ , pH 8.3) with 100  $\mu\text{M}$  oxalate. (C, C') Following 20 min of injection of 10 mg/L humin, the adsorbed particles do not exhibit obvious differences in the presence and absence of oxalate, suggesting that oxalate molecules do not promote the adsorption of humin particles.