Supporting Information

For

Direct Observation of Simultaneous Immobilization of Cadmium and Arsenate at the Brushite-Fluid Interface

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SI TABLES (S1-S6)

SI FIGURES (S1-S9)

Table S1. Aqueous conditions used in $CdCl_2$ solutions.

pH	[$CdCl_2$] (μM)	[Na_2HAsO_4] (μM)	[NaCl] (mM)
5.5-6.0	0	0	0
4.0	5	0	0
6.0	5	0	0
	50	0	0
	500	0	0
8.0	5	0	0

Table S2. Aqueous conditions used in Na_2HAsO_4 solutions.

pH	[$CdCl_2$] (μM)	[Na_2HAsO_4] (μM)	[NaCl] (mM)
4.0	0	50	0
6.0	0	50	0
8.0	0	5	0
	0	50	0
	0	500	0
10.0	0	50	0

Table S3. Aqueous conditions used in the presence of NaCl.

	[$CdCl_2$] (μM)	[Na_2HAsO_4] (μM)	[NaCl] (mM)
Cd	5	0	0.01
pH	5	0	0.1
6.0	5	0	1
	5	0	10
	5	0	100
As	0	50	0.01
pH	0	50	0.1
8.0	0	50	1
	0	50	10
	0	50	100

Table S4. Aqueous conditions used in the presence of both Cd and As.

pH	[$CdCl_2$] (μM)	[Na_2HAsO_4] (μM)	[NaCl] (mM)
6.0	5	50	0
8.0	5	50	0

Table S5. Aqueous conditions used in TEM experiments.

pH	$[CdCl_2]$ (μM)	$[Na_2HAsO_4]$ (μM)	[NaCl] (mM)
6.0	500	0	0
8.0	0	500	0
8.0	5	50	0

Table S6. The speciation calculation in 500 μM $CdCl_2$ solutions using the ECOSAT software.

pH	$[Cd(OH)_2]$ (M)	[CdO] (M)	$CdCO_3$ (M)
4.0	0	0	0
4.5	0	0	0
5.0	0	0	0
5.5	0	0	0
6.0	0	0	0
6.0	0	0	0
6.5	0	0	1.3135×10^{-5}
7.0	0	0	4.5104×10^{-4}
7.5	0	0	4.9504×10^{-4}
8.0	0	0	4.9948×10^{-4}

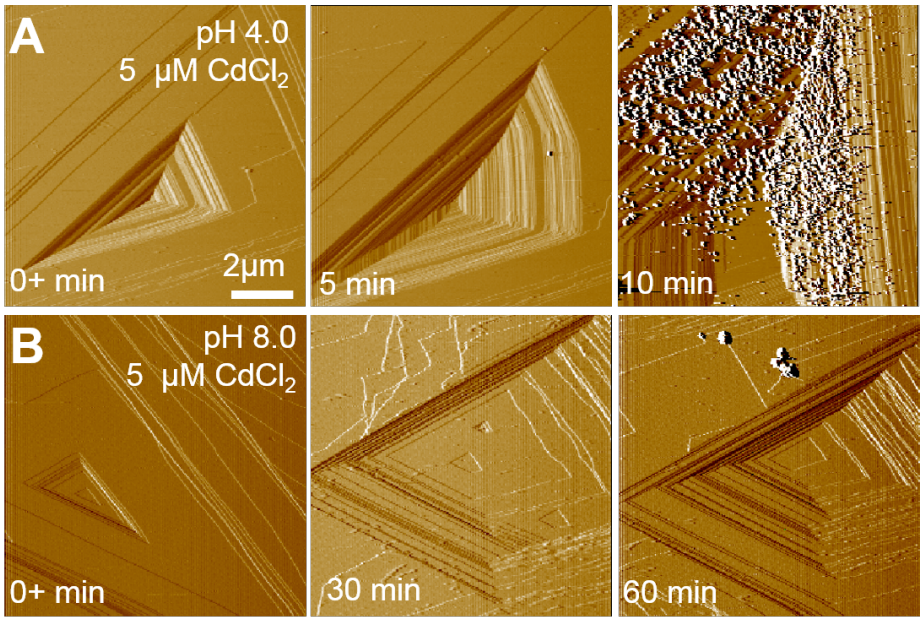


Figure S1. Time sequence of AFM deflection images of brushite dissolved in the presence of 5 μM $CdCl_2$ at (A) pH 4.0, (B) 8.0.

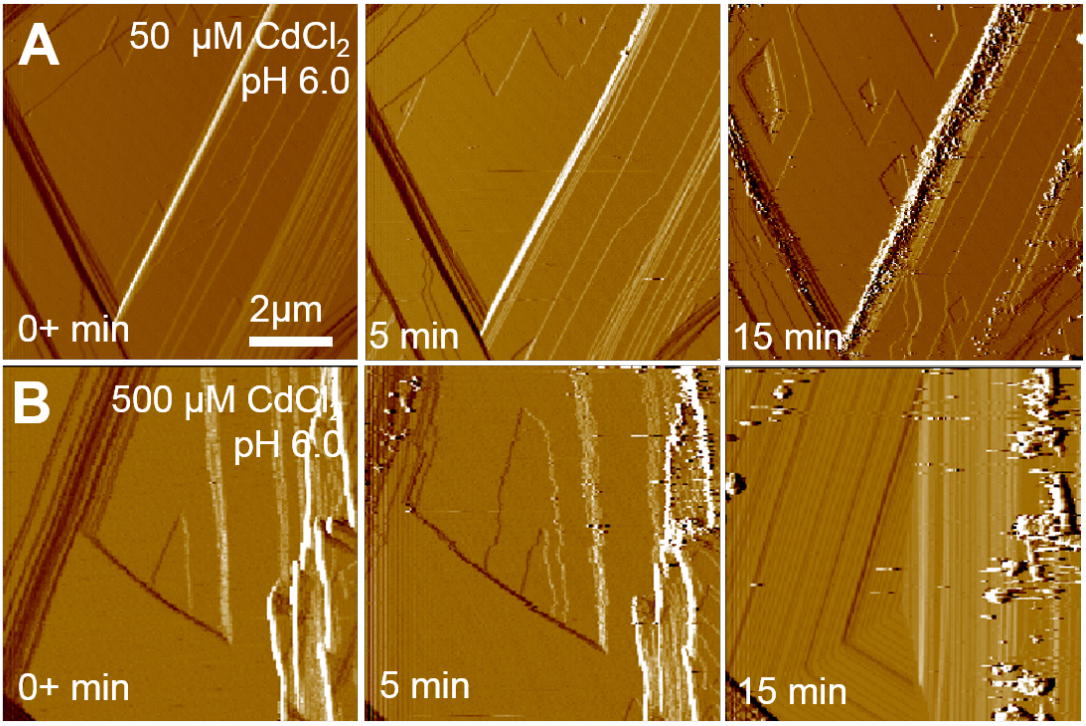


Figure S2. Time sequence of AFM deflection images of brushite dissolved in the presence of (A) 50 μM and (B) 500 μM $CdCl_2$ at pH 6.0.

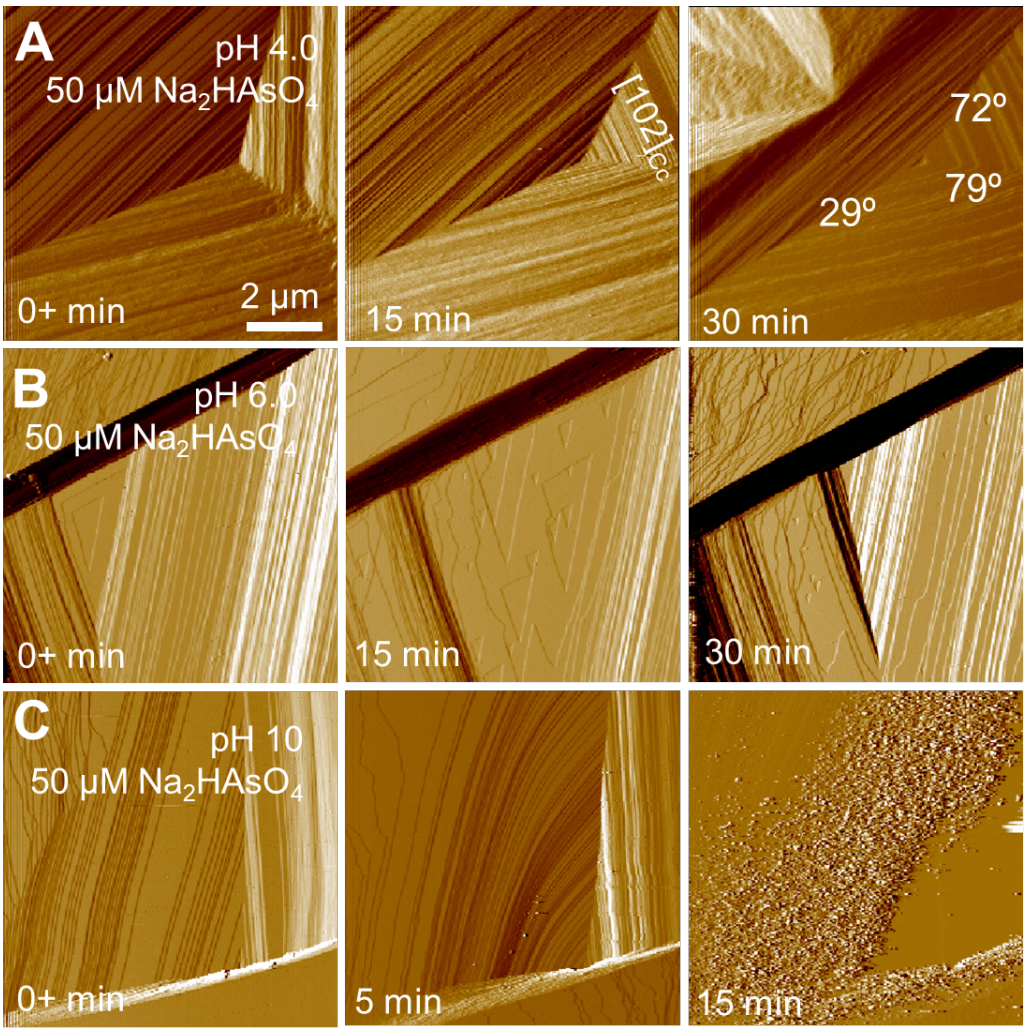


Figure S3. Time sequence of AFM deflection images of brushite dissolved in the presence of 50 µM Na_2HAsO_4 at (A) pH 4.0, (B) 6.0 and (C) 10.0.

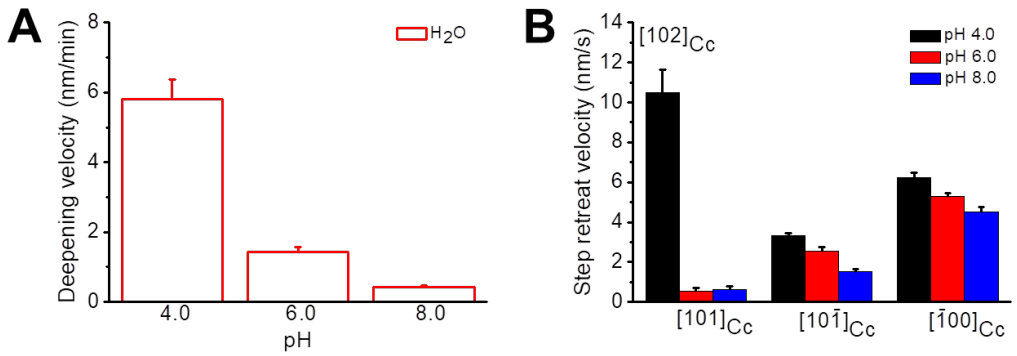
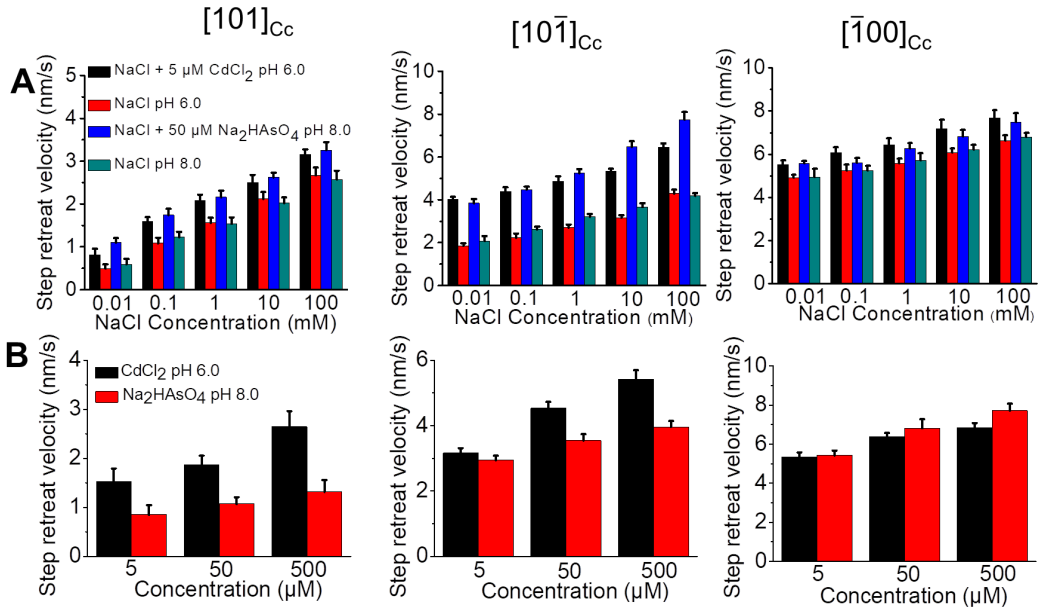


Figure S4. (A) Deepening velocities of etch pits formed at the brushite (010) surface in pure water at pH 4.0, 6.0, or 8.0. (B) Step retreat velocities of the $[102]_{Cc}$, $[101]_{Cc}$, $[10\bar{1}]_{Cc}$, $[\bar{1}00]_{Cc}$ in water at 4.0, 6.0, or 8.0, respectively. All the velocities are presented as mean value ± SD.



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93 **Figure S5.** Step retreat velocities of etch pits along the [101]Cc, [10$\bar{1}$]Cc and

94 [$\bar{1}$00]Cc directions in presence of (A) NaCl + 5 μM $CdCl_2$ or NaCl + 50 μM

95 Na_2HAsO_4, (B) different concentration $CdCl_2$ at pH 6.0 or different concentration

96 Na_2HAsO_4 at pH 8.0, showing that the velocities were changed by the additives. The

97 step retreat velocities are presented as mean value ± SD.

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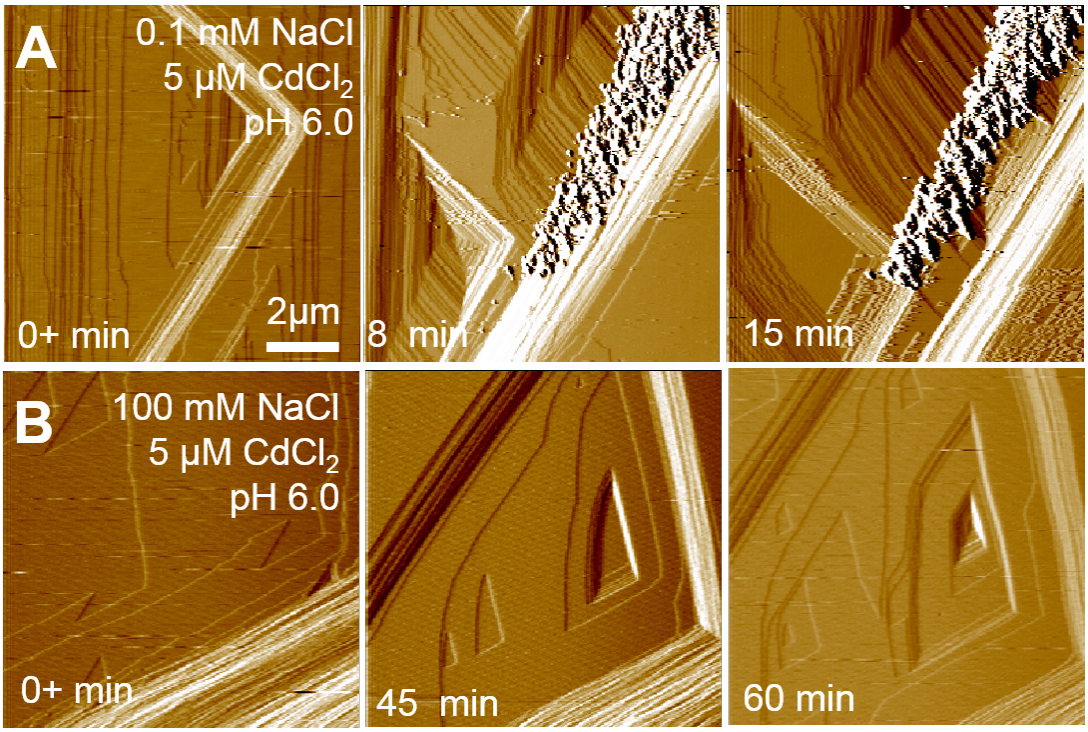


Figure S6. Time sequence of AFM deflection images of brushite dissolved in the presence of 5 μM $CdCl_2$ + (A) 0.1 mM NaCl and (B) + 100 mM NaCl at pH 6.0.

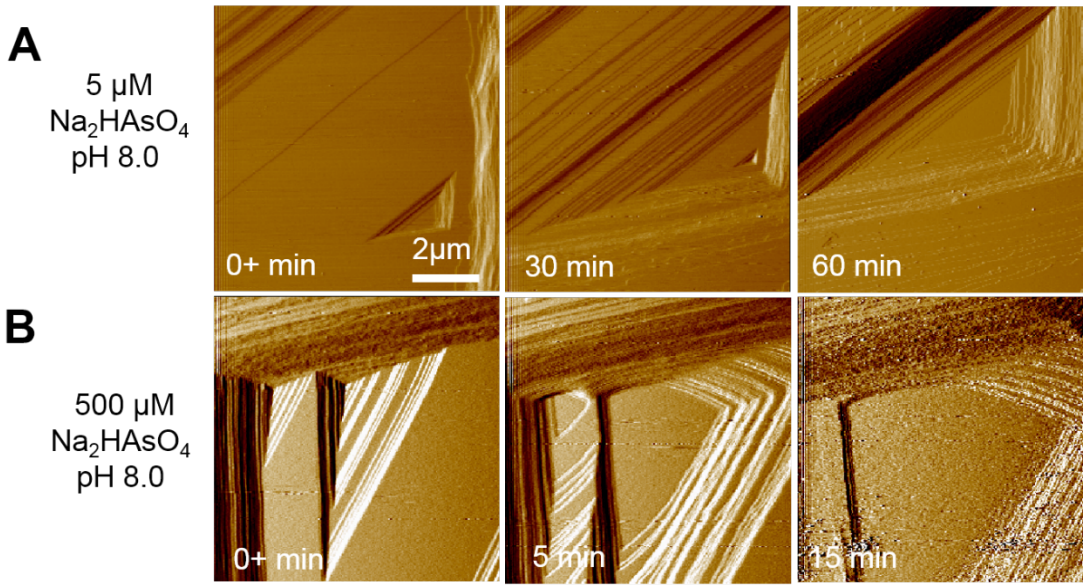


Figure S7. Time sequence of AFM deflection images of brushite dissolved in the presence of (A) 5 μM and (B) 500 μM Na_2HAsO_4 at pH 8.0.

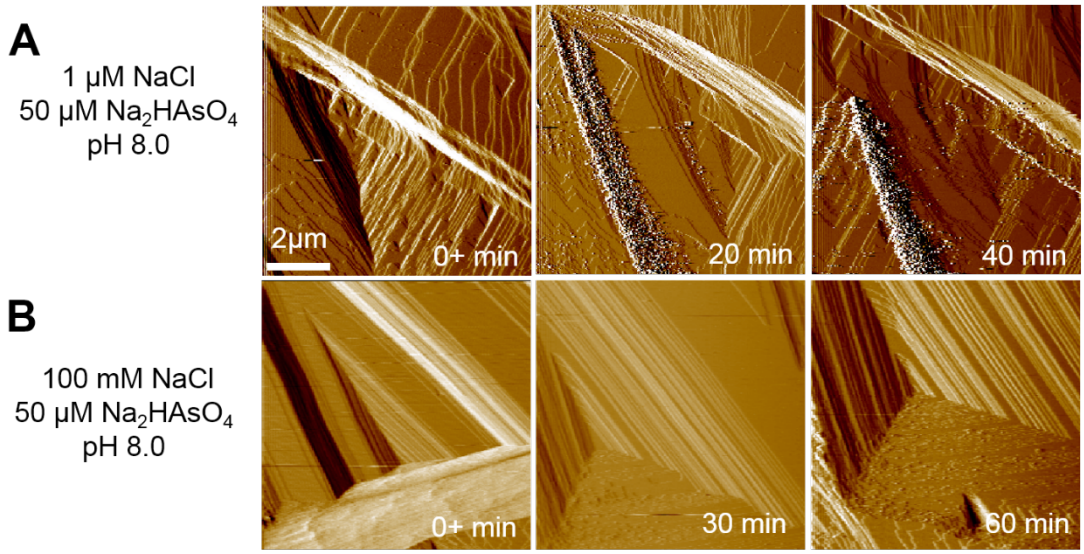
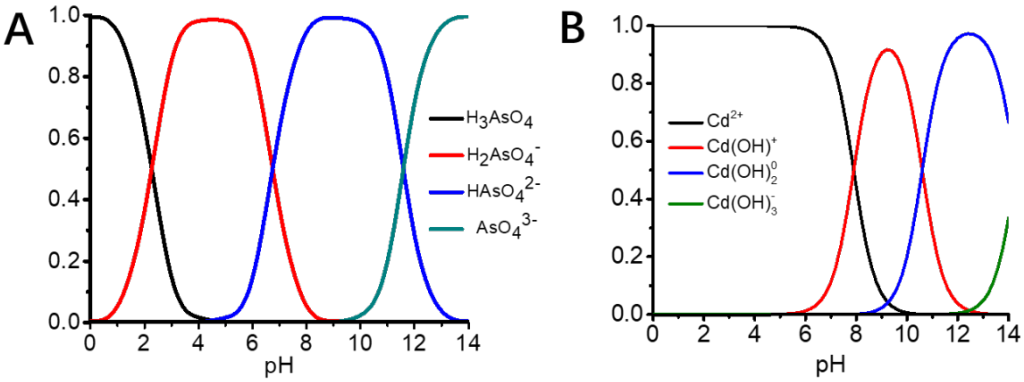


Figure S8. Time sequence of AFM deflection images of brushite dissolved in the presence of 5 µM Na_2HAsO_4 + (A) 0.1 mM NaCl and (B) + 100 mM NaCl at pH 8.0.



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148 **Figure S9.** The relative speciation distributions in (A) As and (B) Cd solutions.

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