Supporting Information for

Nucleation of Aragonite ($CaCO_3$) from Monohydrocalcite ($CaCO_3.H_2O$) Slurries: Kinetics and Activation Energy from Raman Spectroscopy

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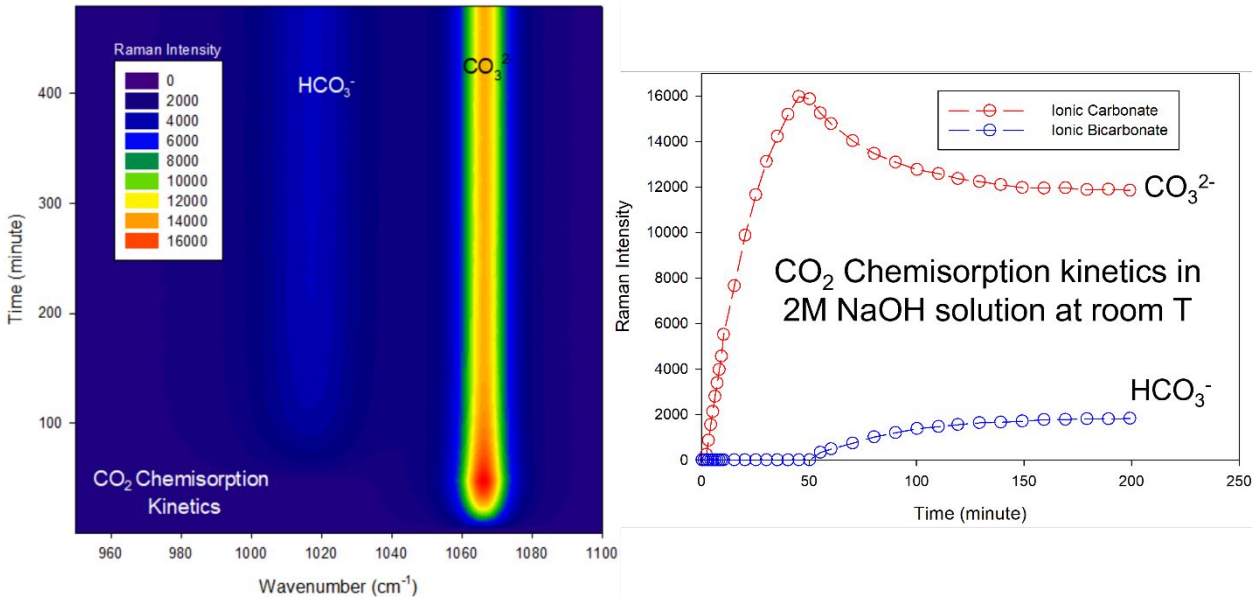


Figure SI-1. CO_2 chemisorption into NaOH solution forming carbonate and bicarbonate ions (carbonate alkaline solution). These kinetic chemical reactions were monitored in real-time by Raman spectroscopy. Case: 25bar of high-purity (99.995%) carbon dioxide were injected in a reactor (autoclave 2L in total) containing 1L de NaOH 2M solution.

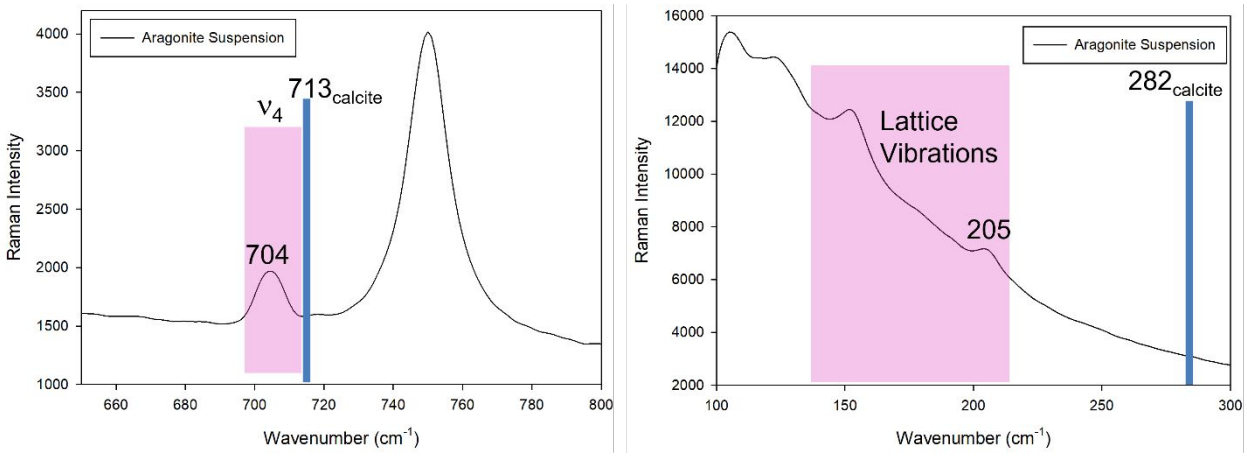


Figure SI-2. Bending and lattice vibration modes allows a simple discrimination between aragonite and calcite. This is fundamental when concurrent formation of aragonite and calcite co-exist because its stretching vibration mode is overlapped for two polymorphs (1085 cm^{-1}).

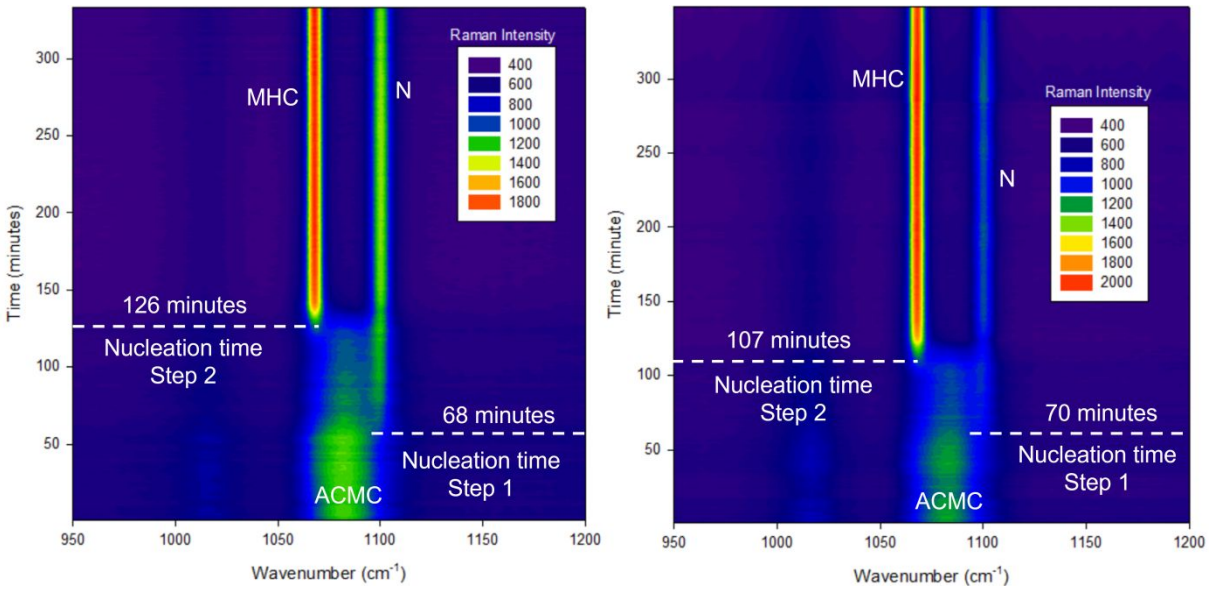


Figure SI-3. Reproducibility of concurrent nucleation of nesquehonite ($MgCO_3.3H_2O$) and monohydrocalcite ($CaCO_3.H_2O$) from amorphous calcium-magnesium carbonate (ACMC) slurry monitored in real-time by Raman spectroscopy. Two additional experiments with respect to Figure 1. ACMC: amorphous calcium-magnesium carbonate; N: nesquehonite; MHC: monohydrocalcite.

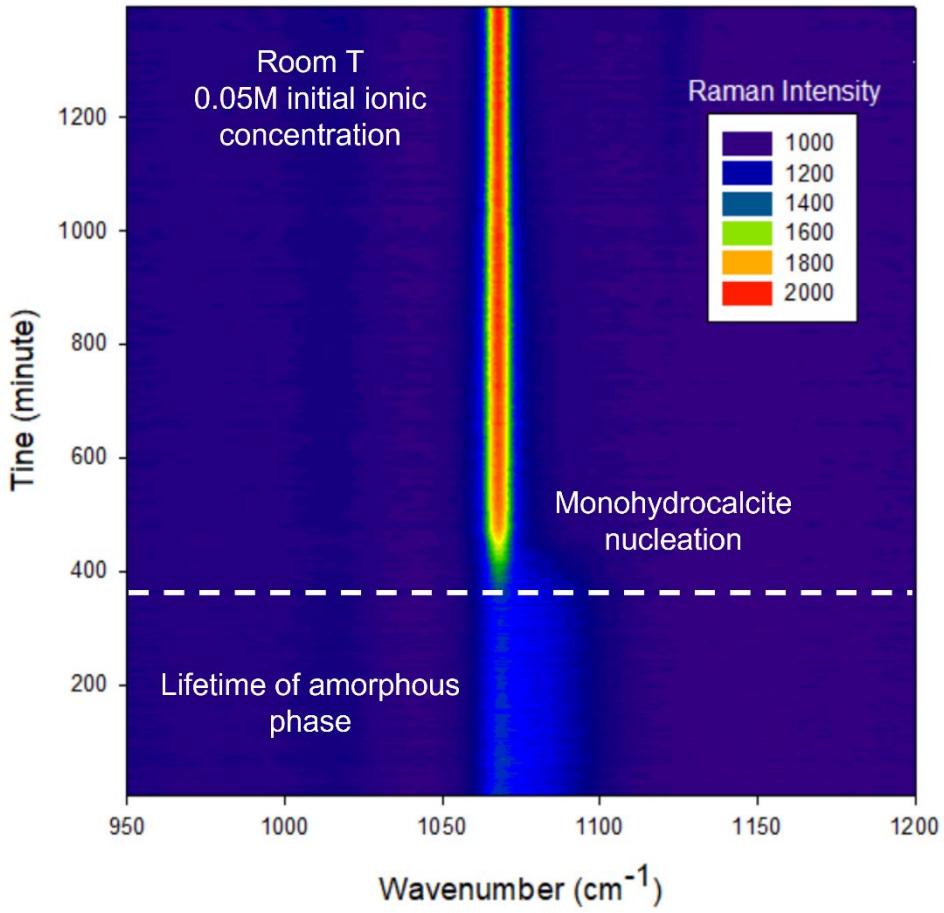
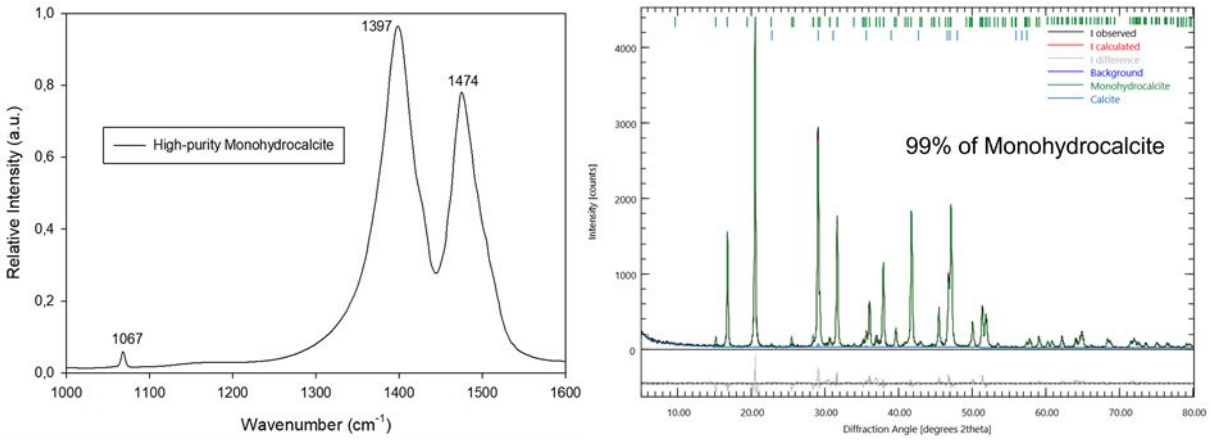


Figure SI-4. Monohydrocalcite nucleation from amorphous calcium-magnesium carbonate monitored by Raman spectroscopy in real-time. Case: 50 mM of Ca and 50 mM of magnesium. In fact, the lifetime of amorphous phase increases with a decrease of ionic concentration (see also Figure 1 and Figure SI-3). Moreover, at low ionic concentration (C≤50 mM), the nesquehonite precipitation was inhibited at room T (see also Figure 1).



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75 Figure SI-5. High-purity of monohydrocalcite determined by infrared spectroscopy and x-ray

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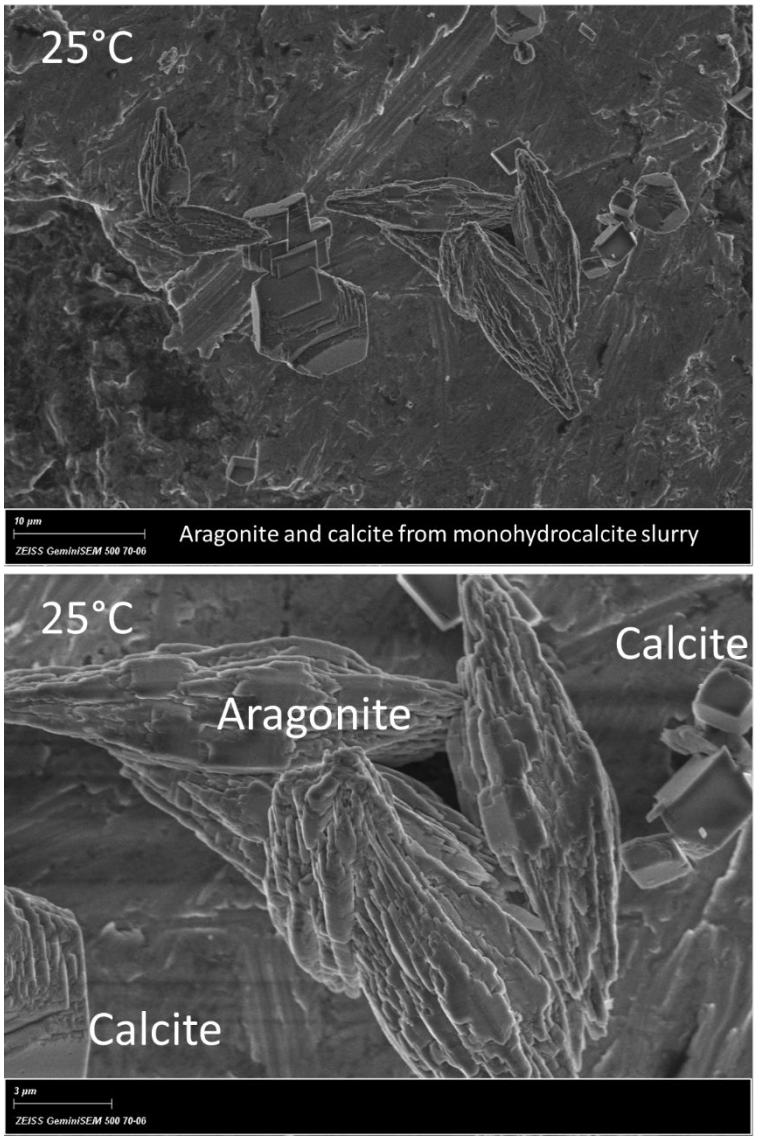

Figure SI-6. FESEM images illustrating the co-existence between aragonite and calcite crystallized from monohydrocalcite slurry at room temperature using dialysis and/or percolating experiments with tap water at room temperature.

Ion	Slope	Intersection	R^2	Time	Date
Calcium	21,8	36,1	0,9995	10:30:51	02/06/2026
Chloride	-48,4	253,6	0,9998	10:30:51	02/06/2026
Potassium	43,7	-4,3	0,9995	10:30:51	02/06/2026
Sodium	43,1	26,9	1	10:30:51	02/06/2026
Ammonium	49,2	100,6	0,9971	10:30:51	02/06/2026
Nitrate	-55	236,9	0,9998	10:30:51	02/06/2026
Magnesium	23,1	23,6	1	10:30:51	02/06/2026
pH	-52,3	360,3	1	10:05:29	11/07/2025

Probe Description	Ca	Cl	K	Na	NH4	NO3	Mg	pH	Units	Time	Date
T=0-F1-R-P-1	0,51	6421	0,14	4235	2,08	86	184	9,51	mg/L	15:34:41	02/06/2026
T=30-F1-R-P-	0,44	801	0,07	430	0,26	20	62	9,64	mg/L	15:31:08	02/06/2026
T=60-F1-R-P-	3,03	216	0,12	90	0,11	10	26	8,48	mg/L	15:27:25	02/06/2026
Eau Robinet	34	4,04	0,04	8,69	0,01	1,36	21	7,7	mg/L	15:23:35	02/06/2026
T=90-F1-R-P-	10	118	0,16	82	0,09	7,54	24	8,32	mg/L	15:20:49	02/06/2026
T=120-F1-R-F	10	140	0,08	93	0,06	7,7	20	8,16	mg/L	15:16:39	02/06/2026
T=F-F1-R-P-1	5,41	289	0,27	105	0,14	12	22	8,44	mg/L	15:13:04	02/06/2026
Eau Robinet	38	4,28	0,05	6,72	0,01	1,13	17	7,84	mg/L	15:09:29	02/06/2026

Eau Robinet = tap water from Grenoble

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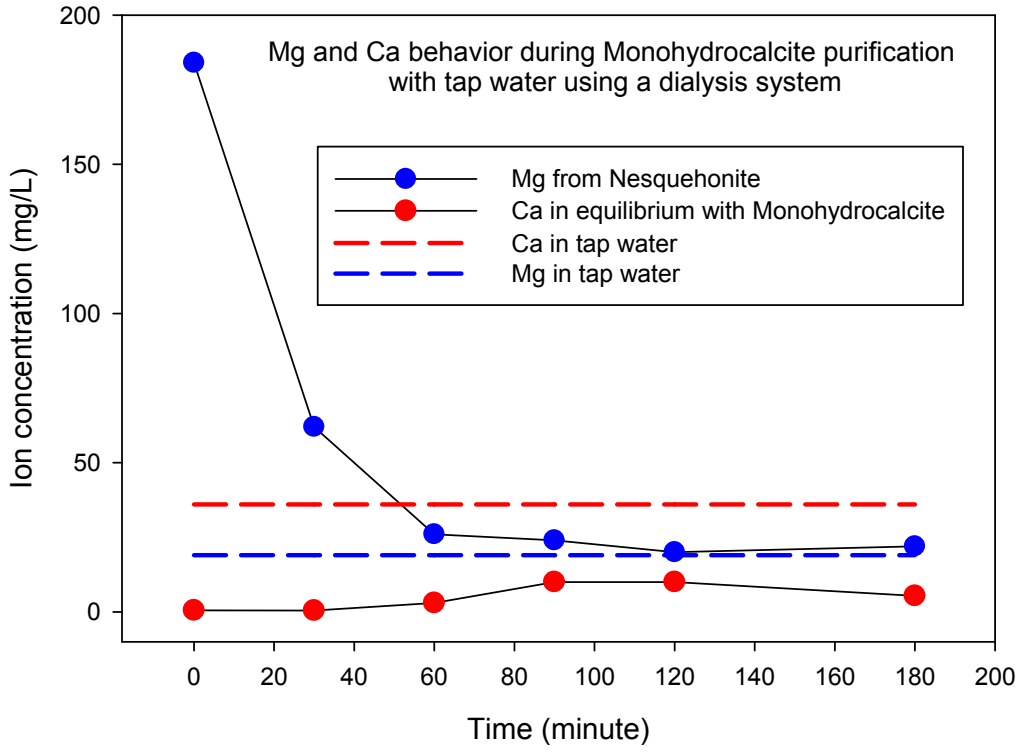

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89 Figure SI-7. Ion concentration and pH in collected-time samples (Imacimus probe) during
90 monohydrocalcite purification with tap water from Grenoble by using a flowthrough reactor (or
91 dialysis technique).

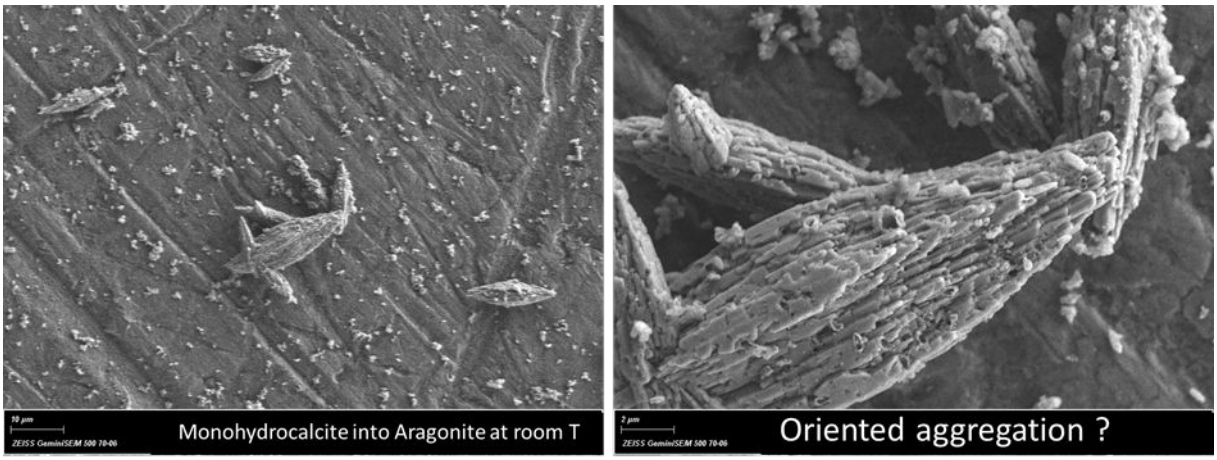

Figure SI-8. FESEM images concerning aragonite aggregates (rice shape) and residual monohydrocalcite (incomplete transformation). This suggesting dissolution-recrystallisation combined with oriented aggregation processes.