Supplementary Information (SI) for Physical Chemistry Chemical Physics.

How non-aqueous media direct the reaction of $Ca(OH)_2$ with CO_2 to different forms of $CaCO_3$: operando mid-infrared and X-ray absorption spectroscopy studies

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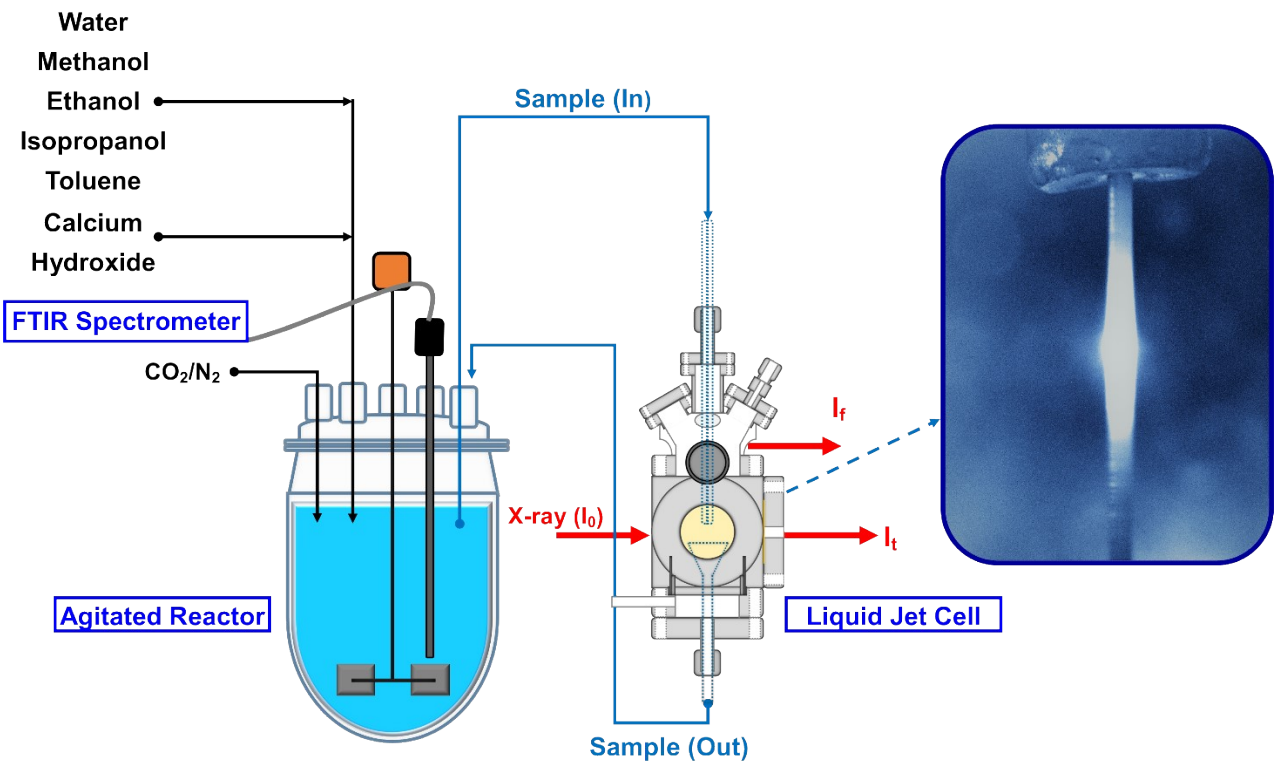

Figure S1. Simplified schematic of the continuous-flow liquid-jet PAT experimental setup for simultaneous *operando* mid-IR and XAS measurements.[1, 2]

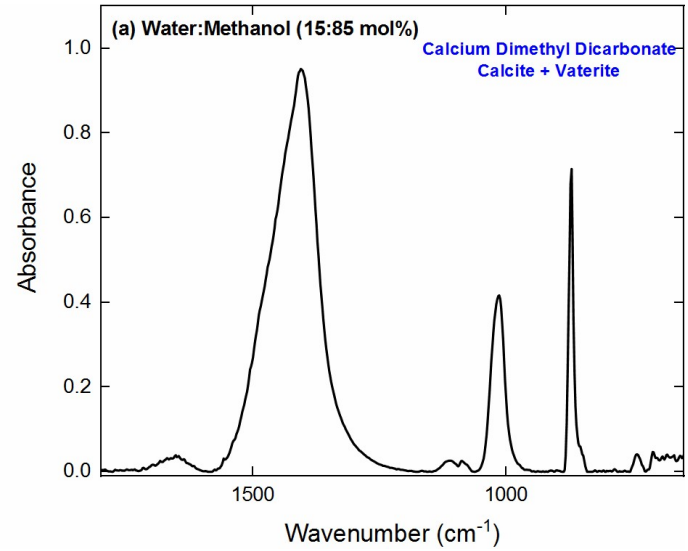

Figure S2. Mid-IR of the aged post-carbonation product for the water-methanol system.

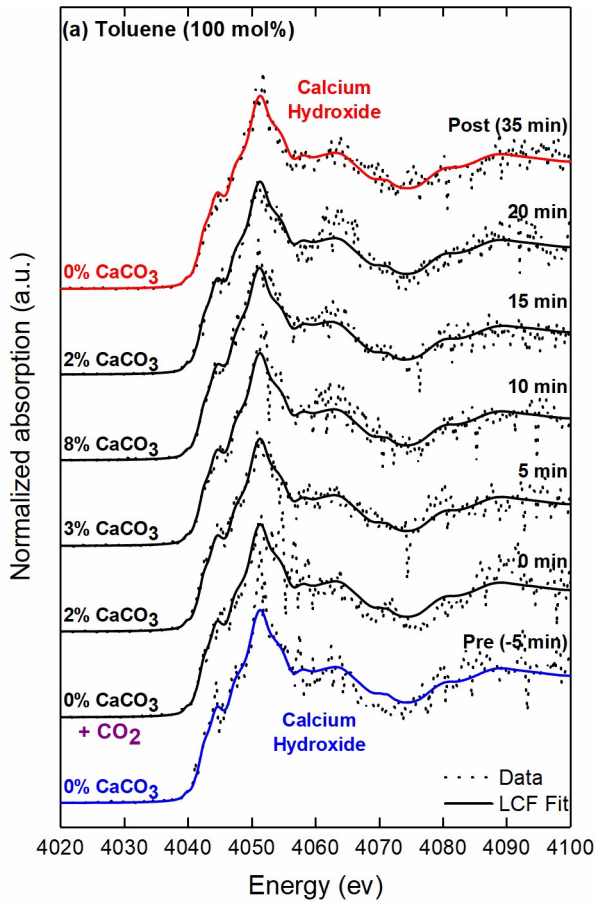

Figure S3. Time-resolved Ca K-edge XANES of the 20-minute carbonation of $Ca(OH)_2$ in pure toluene – no conversion observed.

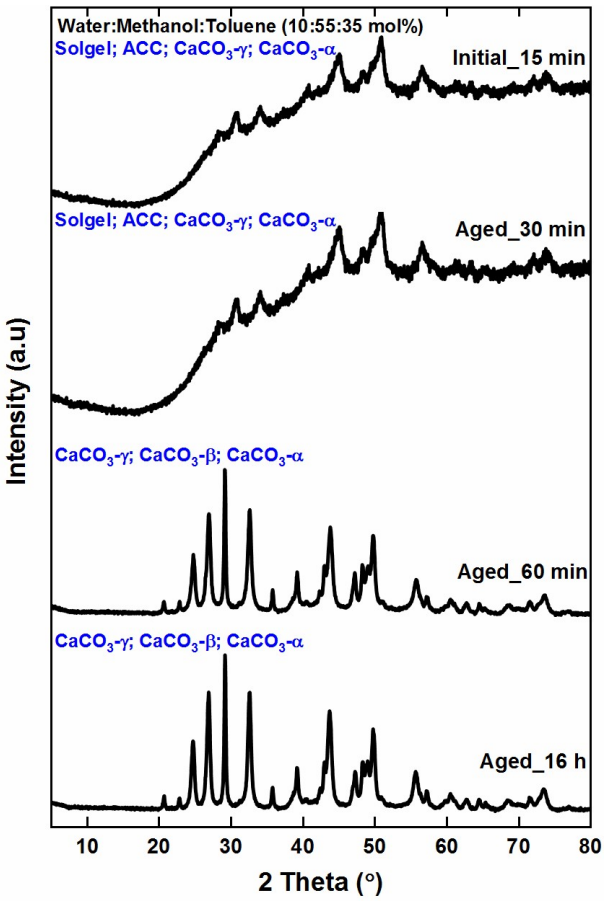

Figure S4. Time-resolved XRD following the aging of the post-carbonation product (15 mins to 16 hours) precipitated in a ternary water-methanol-toluene (10:55:35 mol%) system.

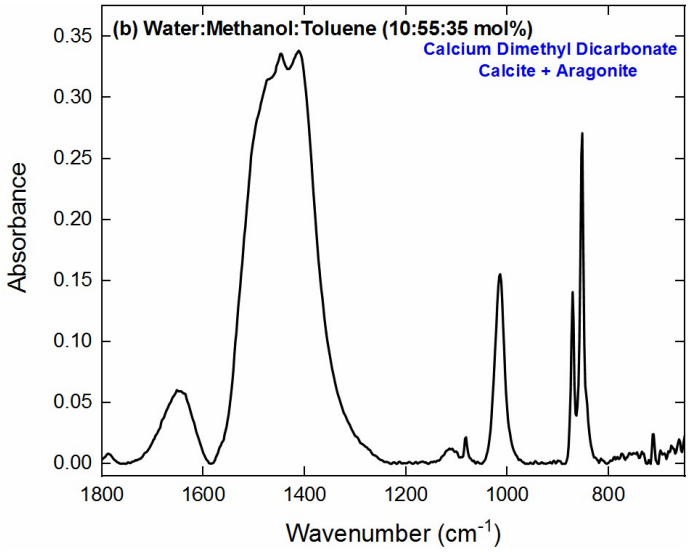

Figure S5. Mid-IR spectrum of the aged (60 hours) post-carbonation products for the water-methanol-toluene system. shows a mixture of calcite and aragonite.

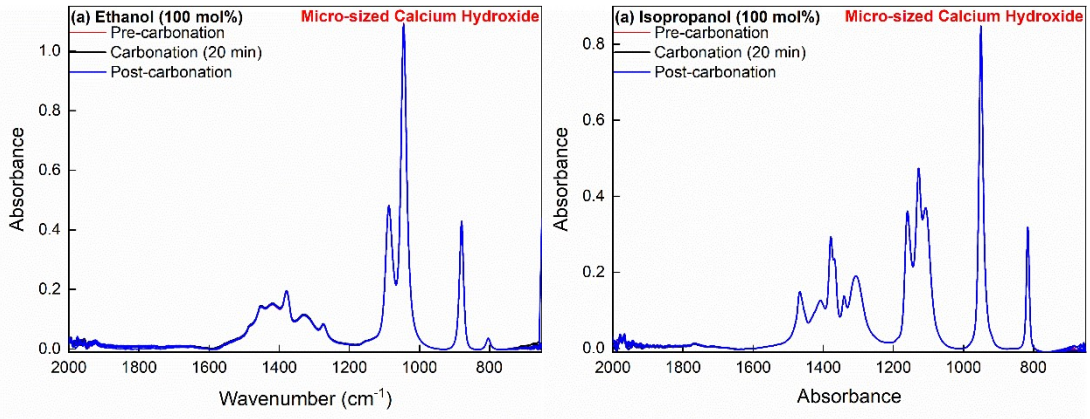

Figure S6. Mid-IR of micro-sized $Ca(OH)_2$ carbonation in pure (a) ethanol (product – no conversion) and (b) isopropanol (product – no conversion).

REFERENCES

1. S. Y. Chang, T. A. Kathyola, E. A. Willneff, C. J. Willis, P. Wilson, P. J. Dowding, G. Cibin, A. B. Kroner, E. J. Shotton and S. L. M. Schroeder, *Reaction Chemistry & Engineering*, 2019, **4**, 679-687.
2. T. A. Kathyola, S.-Y. Chang, E. A. Willneff, C. J. Willis, G. Cibin, P. Wilson, A. B. Kroner, E. J. Shotton, P. J. Dowding and S. L. M. Schroeder, *Industrial & Engineering Chemistry Research*, 2023, **62**, 16198-16206.