

Electronic Supplementary Information (ESI)

Valuable Carbon Nanomaterials Directly Prepared from CO₂ via Sonication in Pure Water

Jungwen Yeh, Yuki Moriya and Masaya Uchida

Contents

Fig. S1. Low-magnification TEM survey panels (representativeness).

Fig. S2. Representative TEM image showing Ca(OH)₂ formation after sonication.

Fig. S3. Atmospheric CO₂ uptake experiment and characterization of the recovered CaCO₃ precursor.

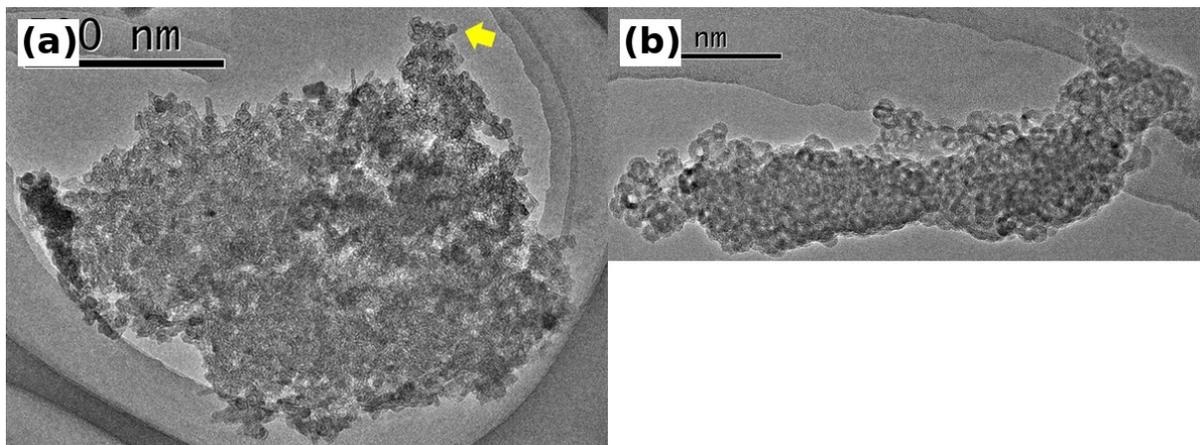
Table S1. Mass balance for the CO₂-uptake control (air vs N₂ headspace).

Table S2. TOC (TC-IC) and NPOC quantification and QA/QC summary.

Table S3. Electrical conductivity (EC) before/after sonication (dataset).

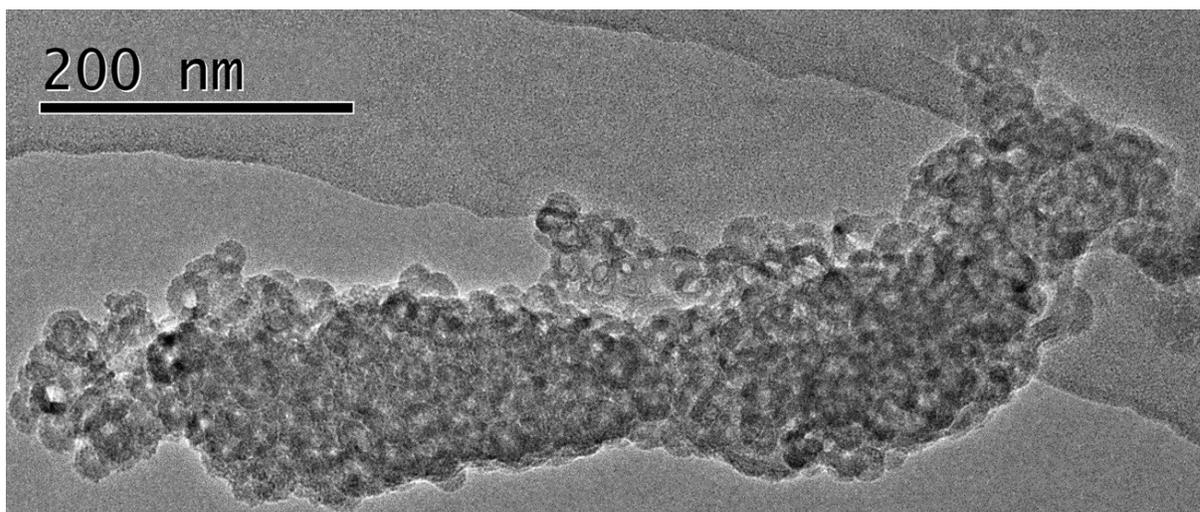
Note S1. Additional discussion of possible side processes (radicals) and limitations.

Fig. S1. Low-magnification TEM survey panels (representativeness).



Caption: Low-magnification TEM survey panels acquired from multiple grids/regions under identical conditions to demonstrate representativeness of the as-made, CNO-enriched mixed carbon. Panel (a) shows CNOs together with minor filamentous (nanotube-like) and sheet-like objects; the yellow arrow highlights a hollow-cored polyhedral particle (cf. Fig. 1a in the main text). Panel (b) shows a region dominated by CNOs. Images are ex situ and are not time-resolved.

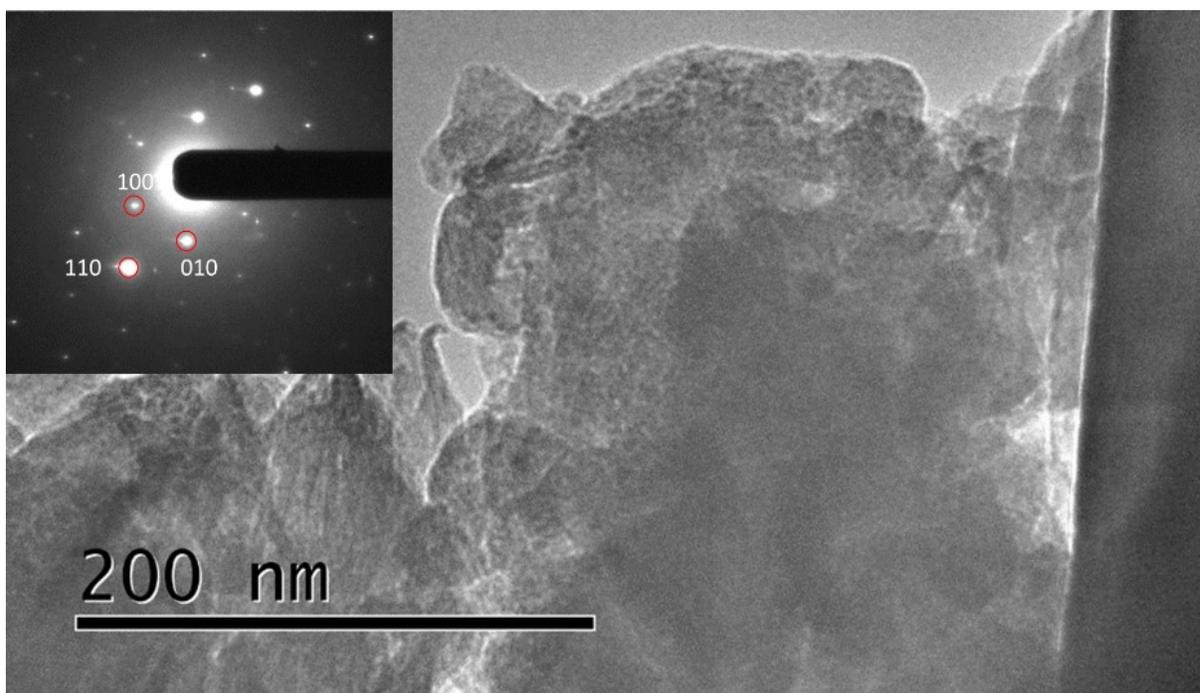
Fig. S2. Evidence for $\text{Ca}(\text{OH})_2$ formation after sonication



Caption: Representative TEM image of solids recovered after sonication, showing plate-like crystallites consistent with $\text{Ca}(\text{OH})_2$ (portlandite).

Fig. S3. Atmospheric CO_2 uptake experiment

Protocol summary (for Fig. S3): $\text{Ca}(\text{OH})_2$ -saturated aqueous solutions were aged for 4 days at room temperature under either ambient air (open to air) or an N_2 headspace (CO_2 -depleted, sealed). After aging, any solids were collected by light centrifugation, rinsed with deionized water, dried, and analyzed by TEM and selected-area electron diffraction (SAED).



Caption: Characterization of solid recovered under ambient air. Representative TEM images and SAED patterns of the solid obtained after 4-day pre-equilibration of a $\text{Ca}(\text{OH})_2$ -saturated solution under ambient air. SAED from single particles shows single-crystal diffraction spots consistent with aragonite CaCO_3 ; no calcite features were observed in the examined regions. Scale bars as indicated.

Table S1. Mass balance for CO₂-uptake control

Condition	Aging time	Recovered solid	Note
Ambient air	4 days	≈34 mg (CaCO ₃)	Carbonate precursor formed in open air.
N ₂ headspace (CO ₂ -depleted)	4 days	trace / negligible (unquantifiable)	Suppressed precursor formation.

Table S2. TOC (TC–IC) and NPOC quantification and QA/QC

Analyte	Value (mg C L ⁻¹)	LOD (mg C L ⁻¹)	LOQ (mg C L ⁻¹)	Note
TC (total carbon)	23.23	0.04	0.12	680 °C catalytic oxidation–NDIR (TOC-V).
IC (inorganic carbon)	8.47	0.006	0.019	Acidification–NDIR (TOC-V).
TOC (TC – IC)	14.75	—	—	Computed from same-lot TC and IC.
NPOC	14.83	0.05	0.16	Acidify (1 M HCl; 1.5%) + sparge 1.5 min.

Note: TOC is reported for clarified supernatants; values represent lower-bound carbon yields because insoluble/retained fractions were not recovered by filtration or acid leaching.

Carbon quantification by TC–IC (TOC) and NPOC. TOC/TC–IC/NPOC measurements were performed by Shimadzu Techno-Research, Inc. (Japan). Liquid samples were handled in glass vials with fluoropolymer-lined caps; headspace was minimized, samples were kept cold (approx. 4 °C), and analyses were completed within 48 h. Total carbon (TC) and inorganic carbon (IC) were measured on the same lot, and TOC was calculated as TOC = TC – IC. Non-purgeable organic carbon (NPOC) was measured on the same lot after acidification to pH ≤ 2 and gas purging to remove IC and purgeable organics. Detection was based on catalytic high-temperature oxidation (approx. 680–1000 °C) to CO₂, followed by NDIR (or coulometry), depending on the instrument configuration. Clarified supernatants were prepared by light centrifugation (see Table S2 for RCF/rpm). LOD/LOQ, calibration, blanks, laboratory control samples (LCS), replicates (RSD), and acceptance criteria are summarized in Table S2.

Yield calculations. Carbon mass m_C (mg) = TOC (mg C L⁻¹) x V_{sup} (L). Percent yield (vs input CaCO₃) = 100 x m_C / $m_{CaCO_3,in}$. Percent C-conversion (vs carbon in CaCO₃) = 100 x m_C / (0.12 x $m_{CaCO_3,in}$).

Table S3. Electrical conductivity dataset

Condition	EC ($\mu\text{S cm}^{-1}$)	Temperature	Note
Before sonication	31.4	25 °C (compensated)	CaCO ₃ suspension.
After sonication	50.2	25 °C (compensated)	Increase consistent with more dissolved ionic species.

Note S1. Additional discussion

Any discussion of sonochemically generated radicals is presented here as a potential side process; radical yields were not quantified in this study and are not required to describe the acid–base CO₂ capture/precipitation pathway.

Where direct time-resolved evidence is not available, claims are framed as proposed/consistent with observations.