# **Supporting information**

# Switchable Oriented Attachment and Detachment of Calcite Nanocrystals

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# Contents

### Characterization techniques

We analyzed nanograins that were obtained by drying filtered dispersion with XRD (Rigaku MiniFlex II) and SEM (Hitachi S-4700, operated at 1.0–5.0 kV). We observed the morphological evolution of the nanograins in the dispersion by TEM (FEI Tecnai F20, operated at 200 kV). The dispersion was dropped on a copper grid covered with a collodion film for TEM observation.

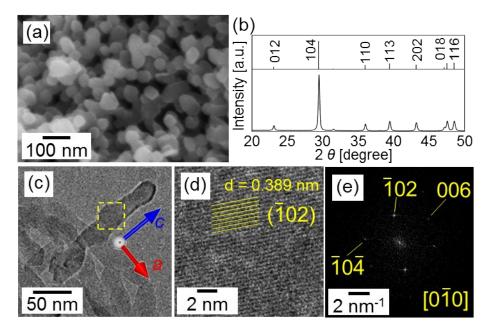
#### Additional information and data

Observation and characterization of calcite nanoblocks after carbonation of Ca(OH)<sub>2</sub> dispersion and elongated calcite nanocrystals formed by oriented attachment of the nanoblocks torough the (001) faces in a basic dispersion (Figure S1)

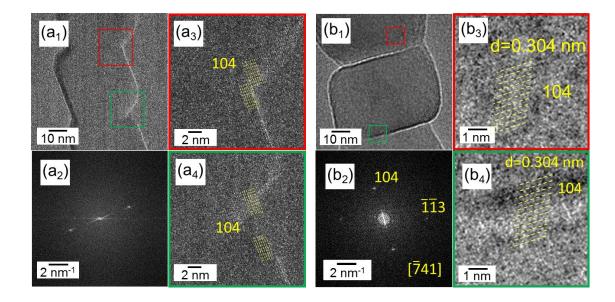
Observation of adjacent calcite nanoblocks arranged in chains in a basic dispersion (Figure S2)

Grain-size distributions of singly dispersed nanocrystals in the initial dispersion after carbonation, and after basification and subsequent neutralization (Figure S<sub>3</sub>)

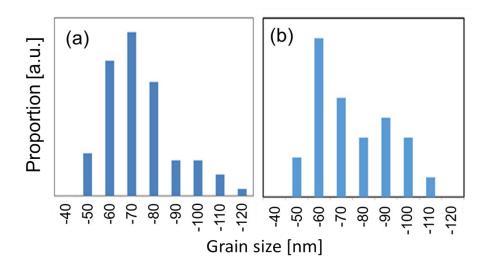
#### Additional information and data



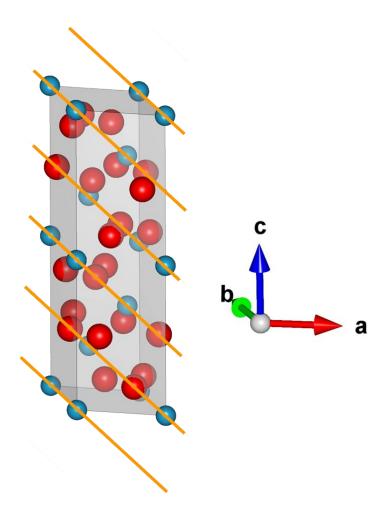
**Fig. S1** A SEM image (a) and a typical XRD pattern (b) of nanocrystals in a dispersion at pH 12 after carbonation; TEM (c) and HRTEM (d) images of elongated nanocrystals in a dispersion maintained at 25°C for 720 h and the FFT pattern (e) of the lattice in the HRTEM image (d). Reproduced from Ref. 17 with permission.



**Fig. S2** HRTEM images (a1, 3-4, b1, 3-4) and FFT patterns (a2, b2) of adjacent calcite nanoblocks in chains in a basic dispersion (pH 12) kept for 120 h.



**Fig. S3** Grain-size distributions of nanocrystals in the initial dispersion at pH 7 after carbonation (a) and in a dispersion at pH 7 after basification and subsequent neutralization (b). The size distributions were estimated from 100 grains in TEM images.



**Fig. S4** Schematic illustration of a calcite ionic configuration with crystal orientation vector. Red and blue spheres are the  $CO_3^{2-}$  and  $Ca^{2+}$  ions, respectively. Orange lines indicate the {104} faces of calcite crystal structure.