Supporting Information

Real-Time Observation of Water-Soluble Mineral Precipitation in Aqueous Solution by *In Situ* High-Resolution Electron Microscopy

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S1. The simulation for the thickness measurement of hexagonal sodium sulfate crystal with tilting.

We identify the thickness of precipitates through simulation of the intensity variation of the $(0\bar{1}10)$ diffraction spot of hexagonal sodium sulfate crystals with tilt angle. The simulation in Figure S4c is obtained by a Fourier transform of projected atomic potentials. The atom density distribution is assumed to be Gaussian with FWHM of nominal atomic diameter. We also simulated other distributions but little variation is found in the results. Bravais–Miller (hkil) indices are used in peak labeling. A certain diffraction spot intensity is acquired by the intensities in the reciprocal-space intersecting the tilting plane.

S2. Supporting Figures and Legends

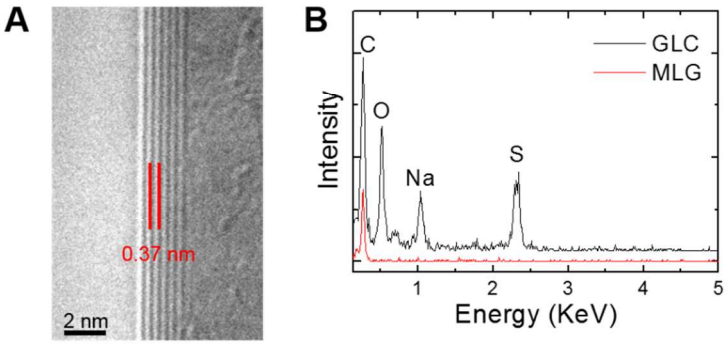

Figure S1. (A) Bright-field TEM image of folded edge for 6-layered graphene sheets. (B) EDS shows Na, S, O, and C (graphene) signals in the graphene pocket containing sodium persulfate aqueous solution (black) and C signal in pristine graphene (red).

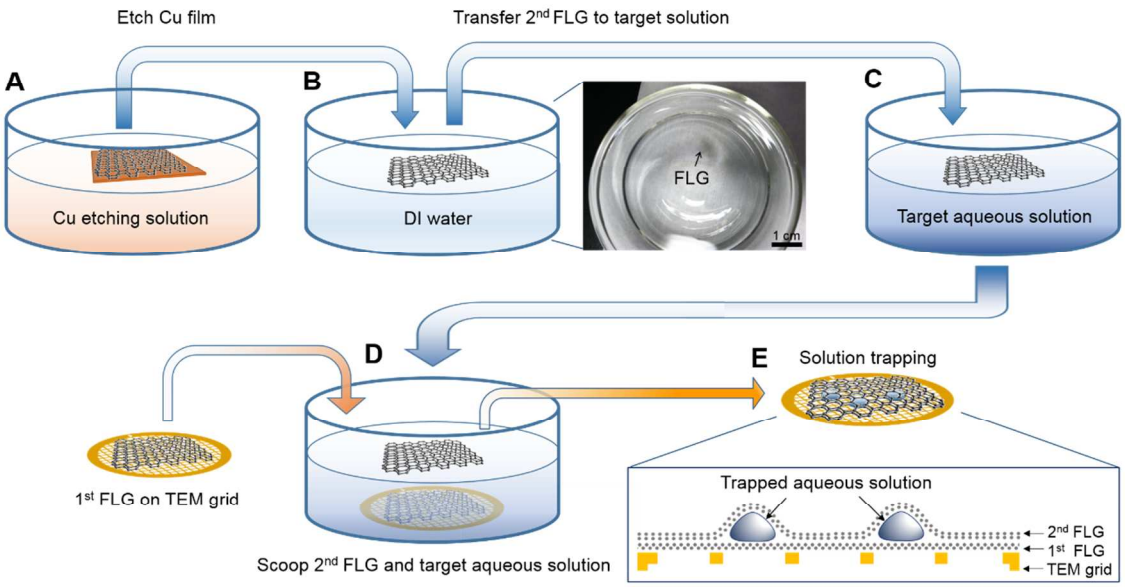

Figure S2. Schematics of GLC preparation flow steps.

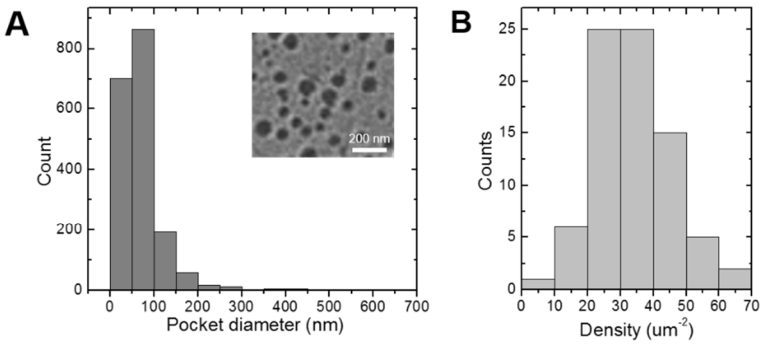

Figure S3. (A) Histogram of graphene pocket size. Inset is bright-field TEM image of typical graphene pockets. (B) Histogram of graphene pocket density.

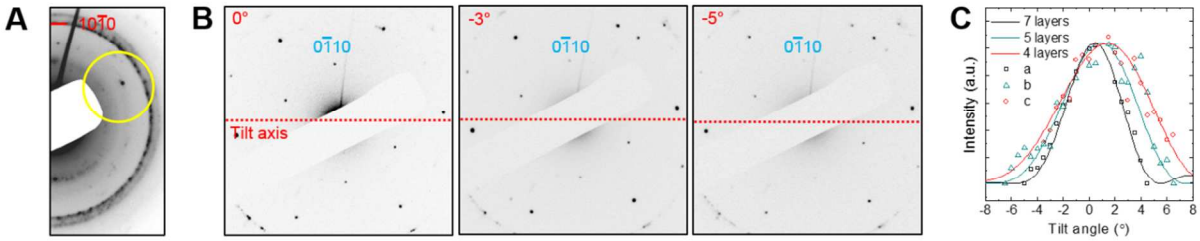

Figure S4. (A) A diffraction pattern shows $(10\bar{1}0)$ diffraction spots from FLG and the other diffraction spots from precipitates. Yellow circle indicates representative position of an objective aperture for DF-TEM imaging in Fig. 1B. (B) Diffraction patterns show intensity change of $(0\bar{1}10)$ diffraction spots of sodium sulfate precipitates tilted by 0°, -3°, and -5°, respectively. (C) Variation of $(0\bar{1}10)$ diffraction intensity of three different precipitates (a, b and c) with tilting angles and simulated lines indicate that the precipitates have 4 - 7 layers thickness, ranging from ~2.9 to 5.1 nm.

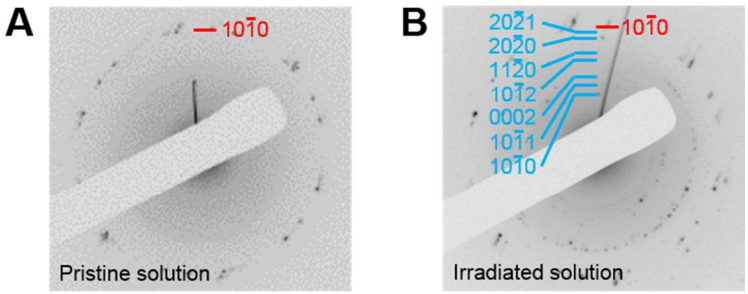

Figure S5. Diffraction patterns show (A) $(10\bar{1}0)$ diffraction spots from FLG before the electron beam irradiation and (B) $(10\bar{1}0)$ diffraction spots from FLG and the other diffraction spots from precipitates after the electron beam irradiation.

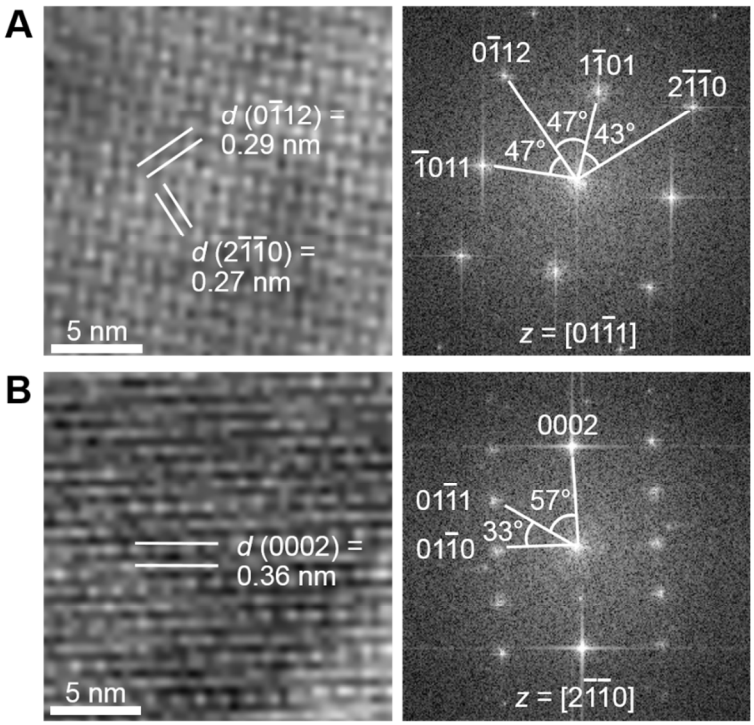

Figure S6. HR-TEM images and the corresponding FFTs of sodium sulfate precipitates with (A) $[01\bar{1}1]$ and (B) $[2\bar{1}\bar{1}0]$ zone axes.

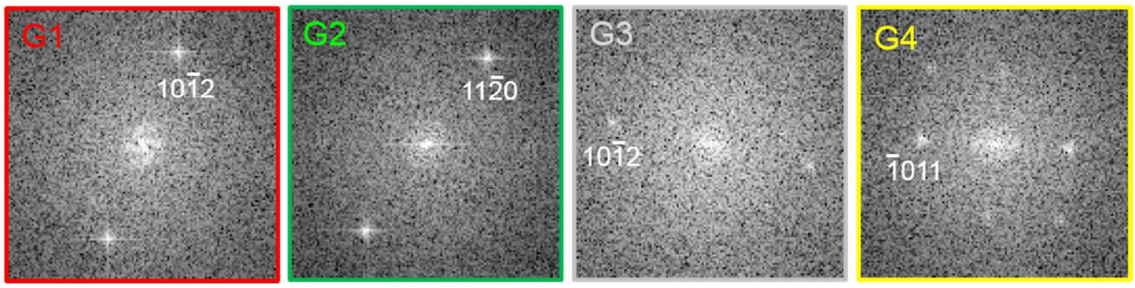

Figure S7. FFTs of G1, G2, G3, and G4 in Figure 3a show crystallographic orientations of each grain. Colors in schematics follow the Figure 3A.

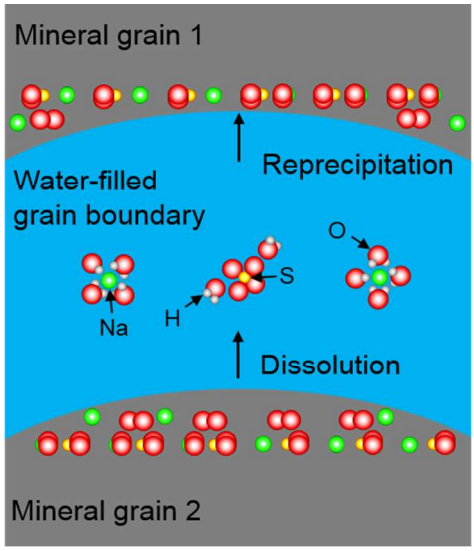

Figure S8. Schematic representation of adatoms on convex surface dissolving in water and then reprecipitation on concave surface to reduce grain boundary area, resulting in grain boundary migration toward center of curvature.

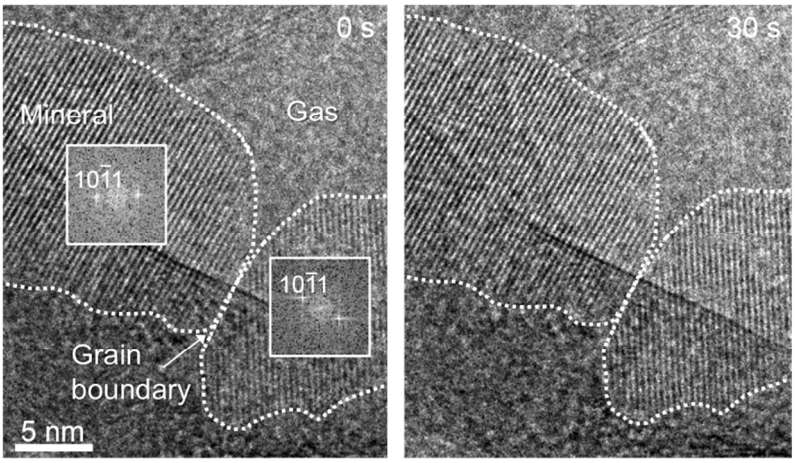

Figure S9. HR-TEM images of sodium sulfate nanocrystals in dried environment. After all water molecules are decomposed to H_2 and O_2 gases, grain boundary is unchanged during observation for 30 sec. Insets in 0 sec show FFTs indicating crystallographic orientations of each grain.

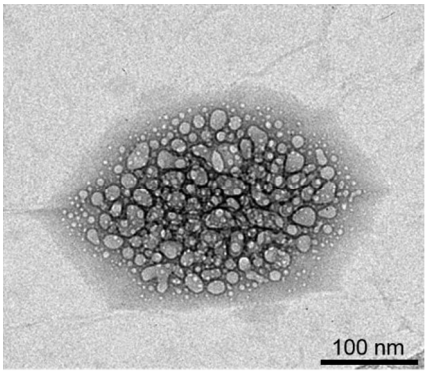

Figure S10. Bright-field TEM image of trapped aqueous solution shows bubbles created by electron beam.

S3. Supporting Movie Legends

Movie S1-S2. Time-lapse dark-field TEM images show the nucleation and growth of precipitates by electron beam irradiation. Movie S1 was acquired with electron beam dose rate of 101 e^-/nm^{2}s, the exposure time of 3 sec and the acquisition time of 5 sec. Snap shots of the Movie S1 are used for Figure 1B. Movie S2 was acquired with electron beam dose rate of 208 e^-/nm^{2}s, the exposure time of 1.5 sec, and the acquisition time of 3 sec. The movies play at 5 Hz.

Movie S3. Time-lapse diffraction patterns show initially $(11\bar{2}0)$ and $(10\bar{1}0)$ diffraction spots from FLG and halo feature from liquid, and then diffraction spots from sodium sulfate precipitates newly appear during electron beam irradiation. Electron beam dose rate was 101 e^-/nm^{2}s, the exposure time was 1 sec and the acquisition time was 3 sec. The movie plays at 5 Hz.

Movie S4. Time-serial HR-TEM images show grain boundary migration and grain rotation during the grain growth of wet sodium sulfate crystals. The motion of three gold nanoparticles with dark contrast indicates that there is a liquid environment. The exposure time was 0.2 sec and the acquisition time was 0.7 sec. The movie plays at 5 Hz. Snap shots of the movie are used for Figure 3A.