

Supplementary Information

# Radiolysis-Induced Crystallization of Sodium Chloride in Acetone by Electron Beam Irradiation

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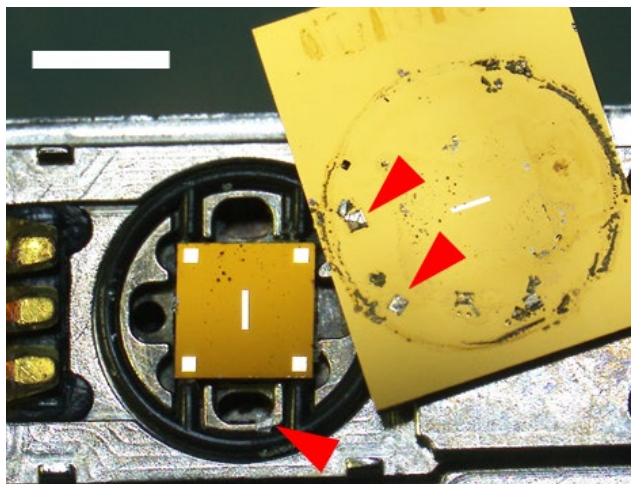
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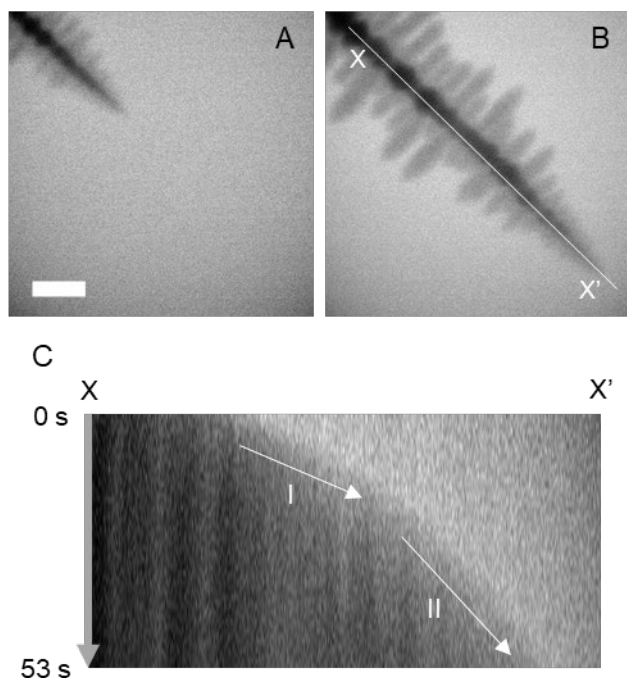
**Supplementary Figures S1–S4**

**Supplementary Table S1**

**Supplementary Videos 1–3**



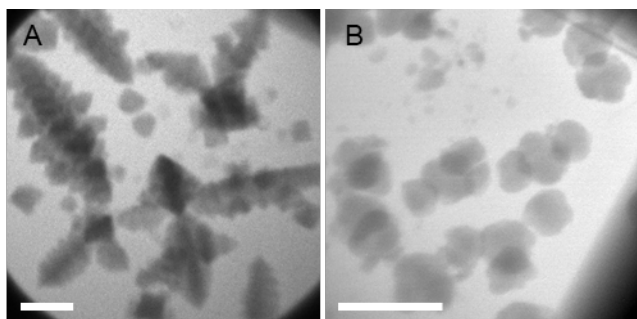
**Supplementary Figure S1.** Crystallization of  $\text{NaClO}_3$  outside the observable view of the liquid cell after mixing of the solutions. The red arrowheads indicate  $\text{NaClO}_3$  crystals. The scale bar (top left-hand corner) is 2 mm.



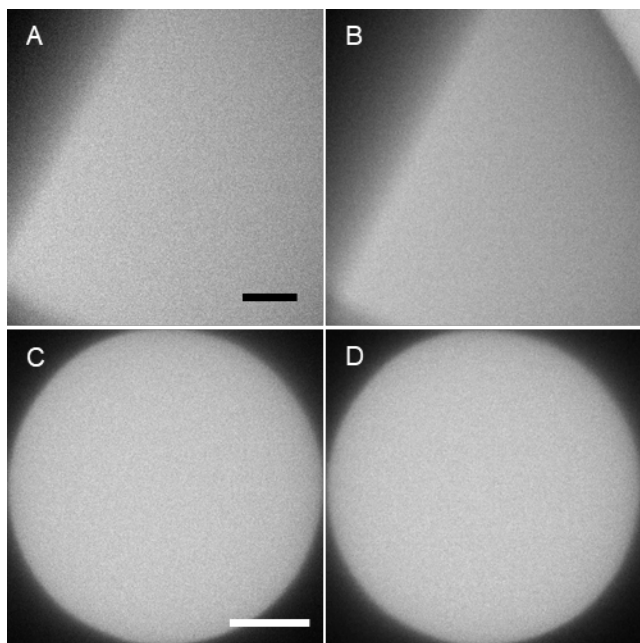
**Supplementary Figure S2.** Measurement of the maximum growth rate of a dendritic crystal.

The crystal was grown during 53 s (A: 0 s, B: 53 s) at an electron dose rate of  $156 \text{ e}^- \text{ nm}^{-2} \cdot \text{s}^{-1}$ .

The displacement of the tip of the stem is visualized by a time–space plot image (C) along the X–X' line in (B), and the slope in this image corresponds to the growth rate. The time–space plot image shows that the growth rate during first 20 s (white arrow I;  $113 \text{ nm s}^{-1}$ ) was larger than that during the subsequent 33 s (white arrow II;  $63 \text{ nm s}^{-1}$ ). Therefore, the maximum growth rate under these conditions was measured from white arrow I as  $113 \text{ nm s}^{-1}$ . The scale bars are  $1 \mu\text{m}$  (A).

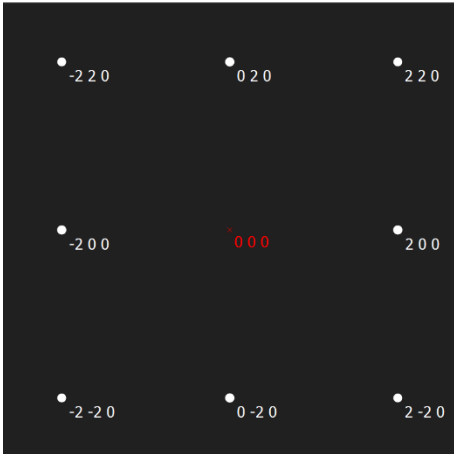
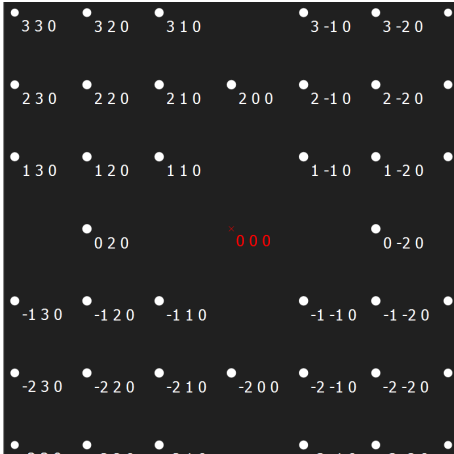


**Supplementary Figure S3.** Various crystals observed in the LC-TEM experiments. (A–B) Rounded crystals formed during the LC-TEM experiments. These rounded crystals formed between dendritic crystals in the mixture of water and acetone solutions (A). The rounded crystals mainly formed in the acetone solution. The electron dose rates were  $18 \text{ e}^- \text{ nm}^{-2} \cdot \text{s}^{-1}$  (A) and  $200 \text{ e}^- \text{ nm}^{-2} \cdot \text{s}^{-1}$  (B). The scale bars are  $2 \mu\text{m}$ .



**Supplementary Figure S4.** Beam effects on saturated aqueous and ethanolic solutions of  $\text{NaClO}_3$ . (A–B) No crystals formed in saturated aqueous  $\text{NaClO}_3$  at an electron dose rate of  $35 \text{ e}^- \text{ nm}^{-2} \cdot \text{s}^{-1}$  during 120 s. Images were captured at about 40 (A) and 120 s (B) after the area had been irradiated by the electron beam. The scale bar is  $1 \mu\text{m}$ , and the scale of (B) is the same as that of (A). The edge of the a-SiN window is visible in the top and bottom left-hand corners (A and B), and a bubble is visible at the top right-hand corner in (B). (C–D) No crystal was formed in saturated ethanolic  $\text{NaClO}_3$  at an electron dose rate of  $2000 \text{ e}^- \text{ nm}^{-2} \cdot \text{s}^{-1}$  during 140 s. Images were captured within 1 s (C) and at 140 s (D) after the area had been irradiated by the electron beam. The scale bar is  $1 \mu\text{m}$ , and the scale of (D) is the same as that of (C).

**Supplementary Table S1.** Crystallographic data and simulated electron diffraction patterns of NaCl and NaClO<sub>3</sub> cubic crystals. The crystallographic data are taken from PDFs provided by the ICDD. The electron-diffraction patterns were simulated by using *ReciPro* software (developed by Dr. Y. Seto). The scales of both simulated patterns are the same, and systematically-absent spots are omitted.

Material	NaCl	NaClO <sub>3</sub>
PDF No.	5-628	5-610
Crystal system	Cubic	Cubic
Symmetry group	Fm $\bar{3}$ m	P2 <sub>1</sub> 3
Lattice constant (Å)	5.6402	6.5756
Simulated [001] SAED Patterns		

**Supplementary Video 1.** Crystallization of dendrites. The video is presented at 2× speed. The scale is  $8.738 \times 8.738 \mu\text{m}$ . Frames of the same event are presented in Figure 2.

**Supplementary Video 2.** Electron-beam-induced nucleation of dendrites. The time and scale are presented in the video. Frames of the same event are presented in Figure 3A–3C.

**Supplementary Video 3.** Electron-beam-induced crystal growth. The time and scale are presented in the video. Frames of the same event are presented in Figure 3D–3F.