

Supporting Information

Demonstrated Gradual Evolution of Disorder in Crystalline Structures between Single Crystal and Polycrystal via Chemical and Physicochemical Approaches

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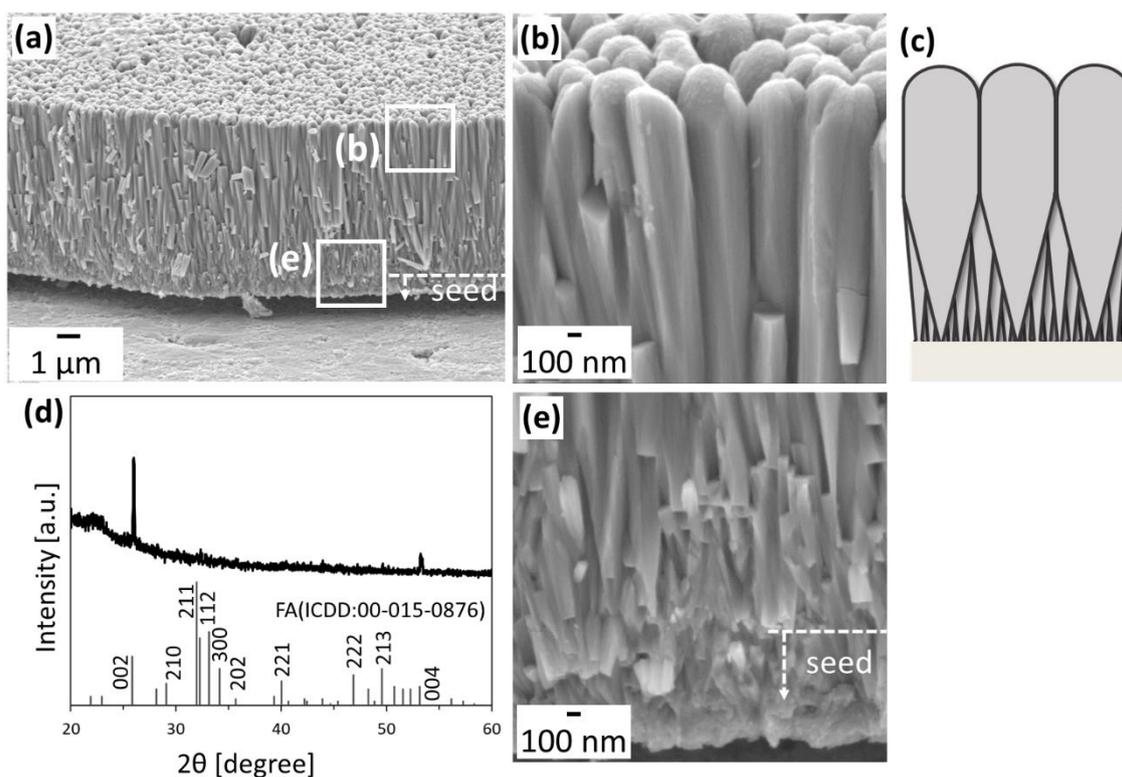


Fig. S1 SEM (a, b, e) and schematic (c) images of a cross-section and a typical X-ray diffraction pattern (d) of c-axis-oriented FA films after subsequent growth in s-SBF1.0 at $[F^-] = 1.50 \text{ mmol/dm}^3$ for 24 h on the seed layer without stirring.¹

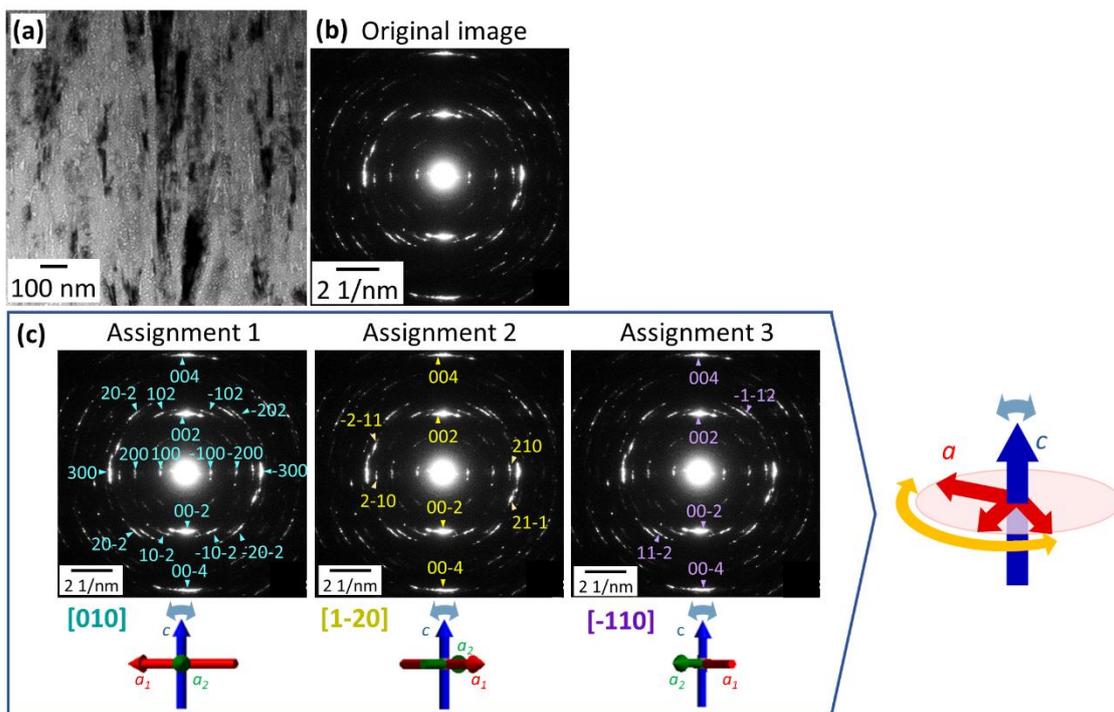


Fig. S2 Assignments of the SAED image (b, c) obtained from a TEM image (a) of c -axis-oriented FA nanorods after subsequent growth in s -SBF1.0 at $[F^-] = 1.50 \text{ mmol/dm}^3$ with addition of acetic acid and without stirring.

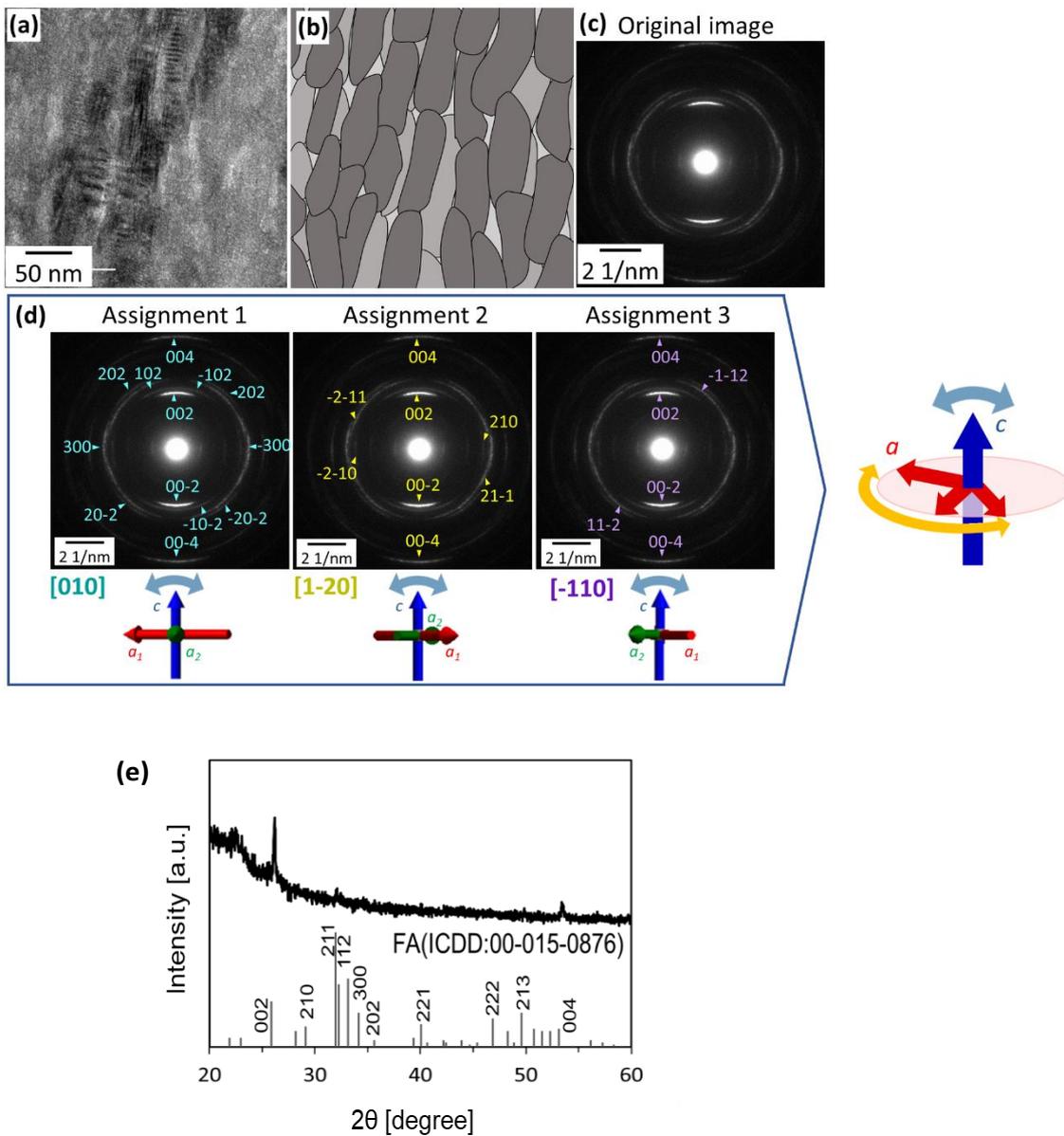


Fig. S3 Assignments of the SAED image (c, d) obtained from the TEM image (a) with a schematic illustration (b) showing a cross-sectional view and a typical X-ray diffraction pattern (e) of *c*-axis-oriented FA nanograins after subsequent growth in *s*-SBF1.0 at $[F^-] = 1.50 \text{ mmol/dm}^3$ with addition of acetic acid and with stirring. The TEM image was obtained from an FIB-cut sample.

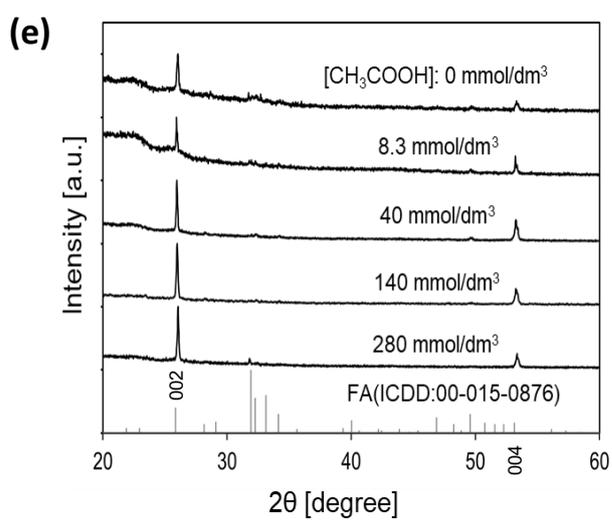
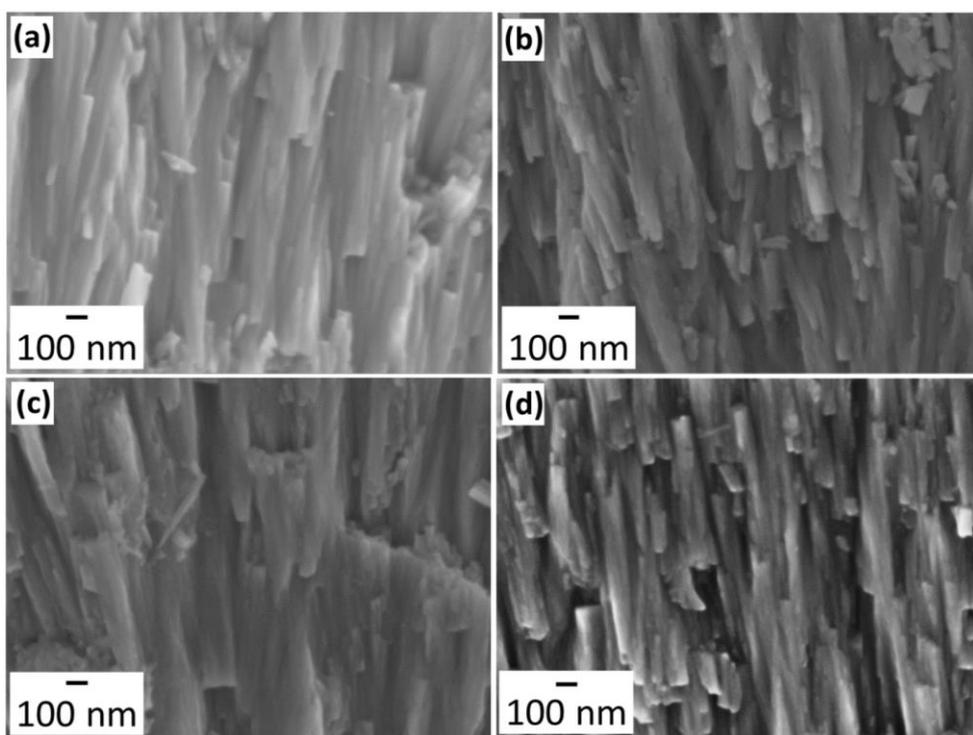


Fig. S4 SEM (a–d) of a cross-section and X-ray diffraction patterns (e) of *c*-axis-oriented FA films on a PVA sheet in *s*-SBF1.0 at $[F^-] = 1.50 \text{ mmol/dm}^3$ with addition of acetic acid and without stirring. (a) $[\text{CH}_3\text{COOH}] = 8.3 \text{ mmol/dm}^3$, (b) $[\text{CH}_3\text{COOH}] = 40 \text{ mmol/dm}^3$, (c) $[\text{CH}_3\text{COOH}] = 139 \text{ mmol/dm}^3$, (d) $[\text{CH}_3\text{COOH}] = 280 \text{ mmol/dm}^3$.

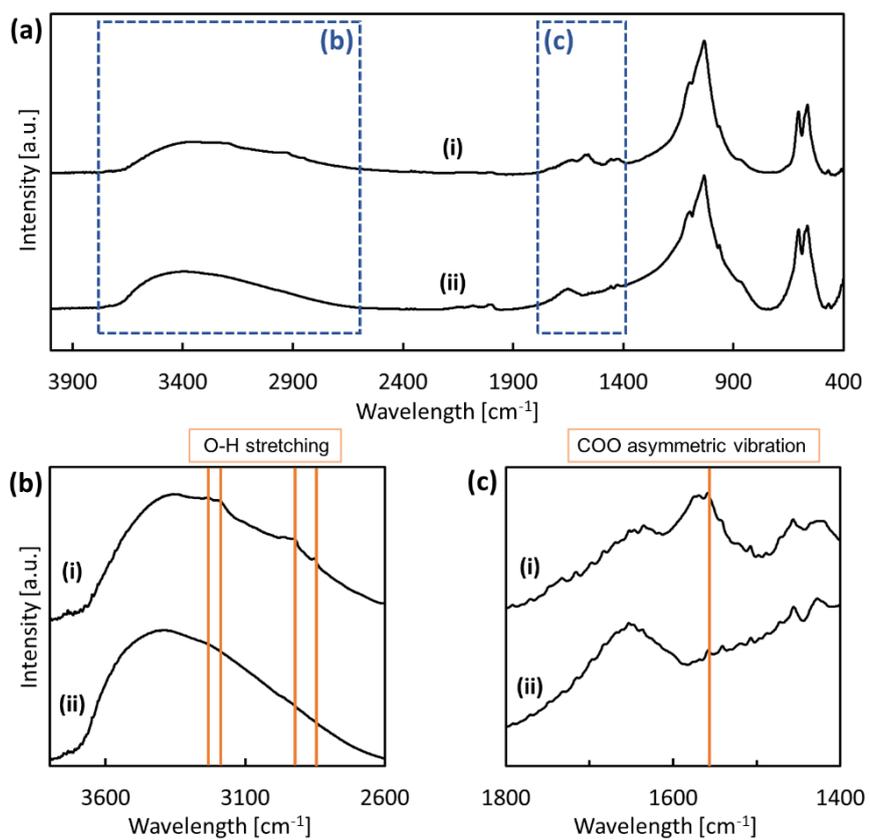


Fig. S5 FT-IR spectra (a–c) of films grown in in s-SBF1.0 at $[F^-] = 1.50 \text{ mmol/dm}^3$ with (i) and without (ii) addition of acetic acid (280 mmol/dm^3).

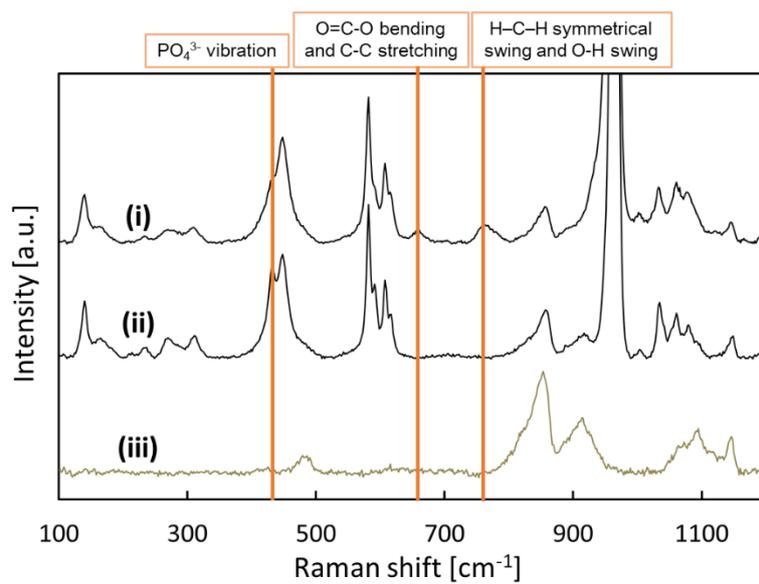


Fig. S6 Raman spectra of films grown in in s-SBF1.0 at $[F^-] = 1.50 \text{ mmol dm}^{-3}$ with (i) and without (ii) addition of acetic acid (280 mmol /dm^3) and a PVA sheet (iii).

Reference

- 1 S. Kanazawa, Y. Oaki and H. Imai, *Nanoscale Adv.*, 2022, **4**, 1538–1544.