

## Supporting Information

### Designed Nanostructures created via Physicochemical Switching of the Growth Mode between Single Crystal and Mesocrystal

Sayako Kanazawa,<sup>a</sup> Yuya Oaki,<sup>a</sup> Hiroaki Imai<sup>a\*</sup>

<sup>a</sup>*Department of Applied Chemistry, Faculty of Science and Technology, Keio University, 3-14-1 Hiyoshi, Kohoku-ku, Yokohama 223-8522, Japan*

\*hiroaki@aplc.keio.ac.jp

Table S1 The width and length of the grains in the films produced under various conditions.

Solution	Stirring	Position	Width(nm)	Length(nm)	Figure
SBF1.0	w/o	upper part	~300	>1 μm	Fig. 2b
SBF1.0	w/o	elementary part	~100	>1 μm	Fig. 2c
SBF2.0	w/o	upper part	~300	>1 μm	Fig. S3b
SBF2.0	w/o	elementary part	~100	>1 μm	Fig. S3c
SBF1.0	w/	upper part	80	500	Fig. 2e
SBF1.0	w/	intermediate part	40	300	Fig. 2f
SBF1.0	w/	elementary part	30	100	Fig. 2g
SBF2.0	w/	upper part	50	200	Fig. S5c
SBF2.0	w/	intermediate part	40	100	Fig. S5d
SBF2.0	w/	elementary part	30	70	Fig. S5e

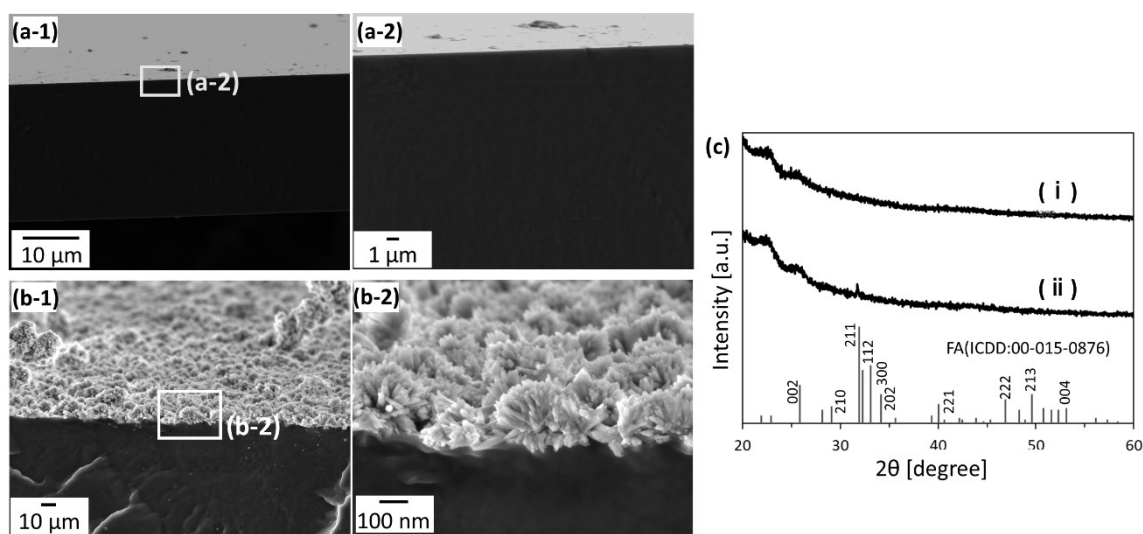


Fig. S1 SEM images of the cross-sectional view of the original PVA sheet (a-1, a-2) and the seed layer after subsequent growth for 6 h in *s*-SBF3.75 at  $[F^-] = 2.25 \text{ mmol dm}^{-3}$  (b-1, b-2). Typical XRD patterns (c) of the PVA sheet (i) and the seed layer (ii).

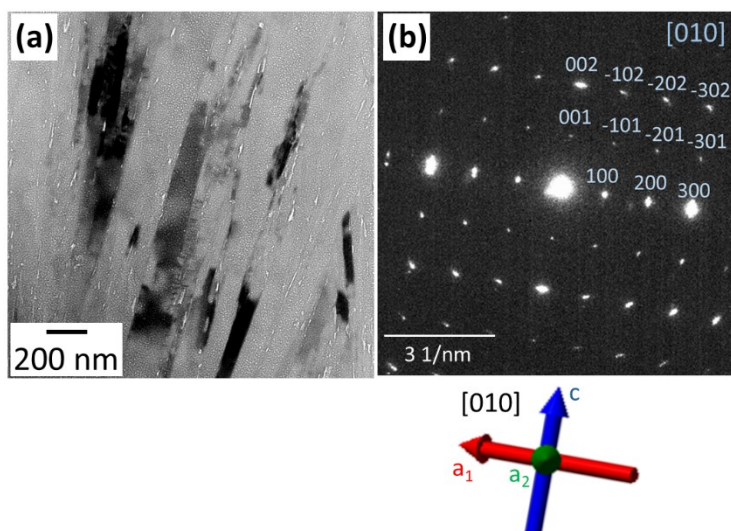


Fig. S2 A typical TEM image (a) and SAED pattern (b) of an FIB-cut plate obtained from the film after subsequent growth without stirring in *s*-SBF1.0 at  $[F^-] = 1.50 \text{ mmol dm}^{-3}$  for 24 h on the seed layer.

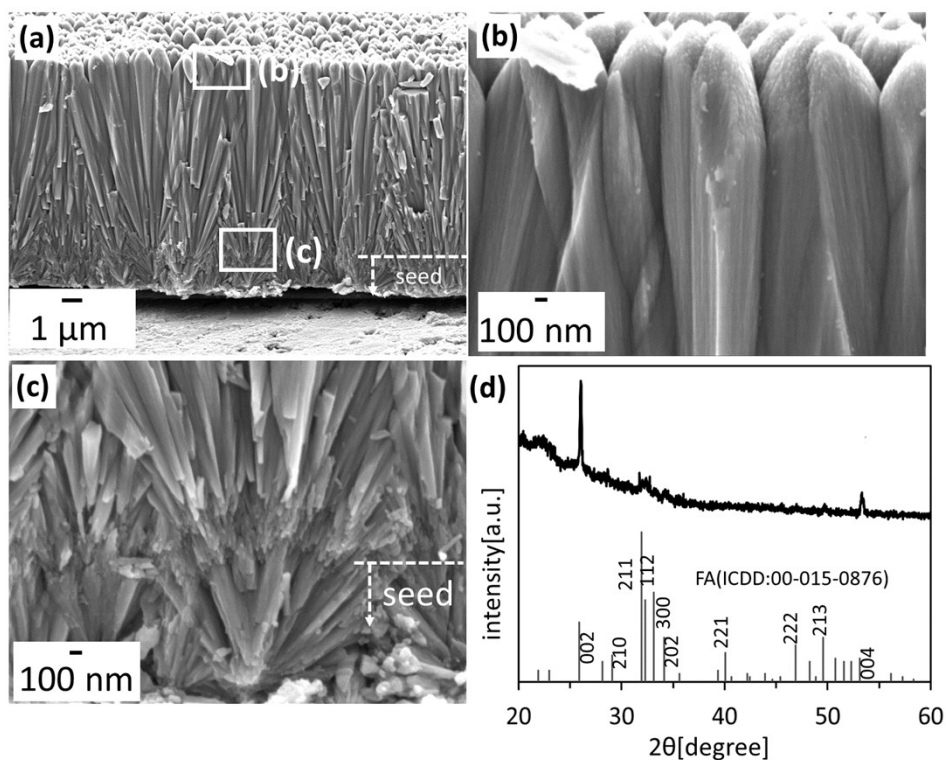


Fig. S3 SEM images (a–c) of the cross-sectional view and a typical XRD pattern (d) of the films after subsequent growth without stirring in s-SBF2.0 at  $[F^-] = 1.50 \text{ mmol dm}^{-3}$  for 24 h on the seed layer.

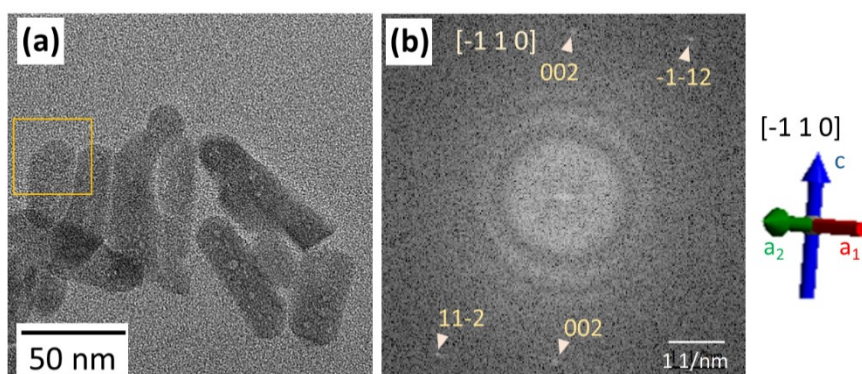


Fig. S4 A typical TEM image (a) and fast Fourier transform (FFT) pattern (b) of grains obtained by fracturing the film after subsequent growth with stirring in s-SBF1.0 at  $[F^-] = 1.50 \text{ mmol dm}^{-3}$  for 24 h on the seed layer.

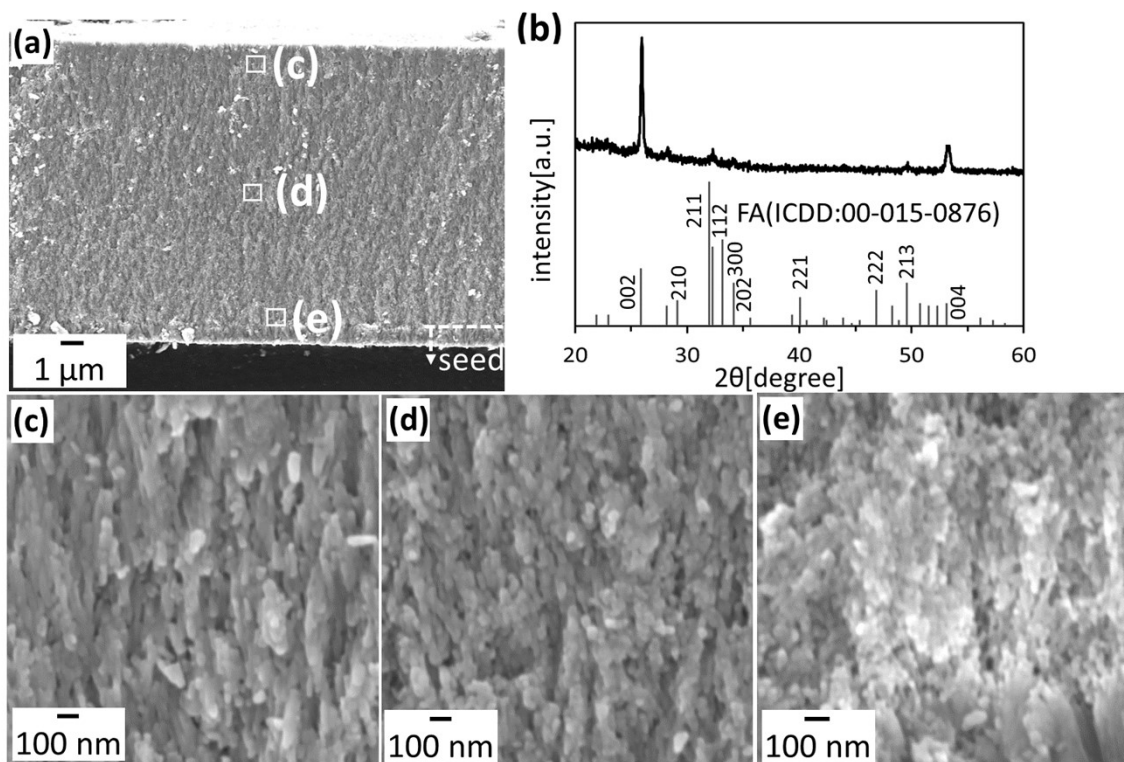


Fig. S5 SEM images (a, c–d) of the cross-sectional view and a typical XRD pattern (b) of the films after subsequent growth with stirring in s-SBF2.0 at  $[F^-] = 1.50 \text{ mmol dm}^{-3}$  for 24 h (a and b) on the seed layer. □

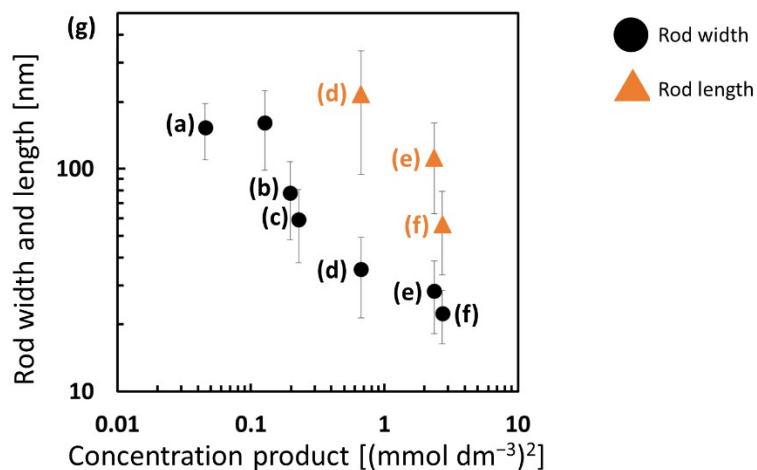
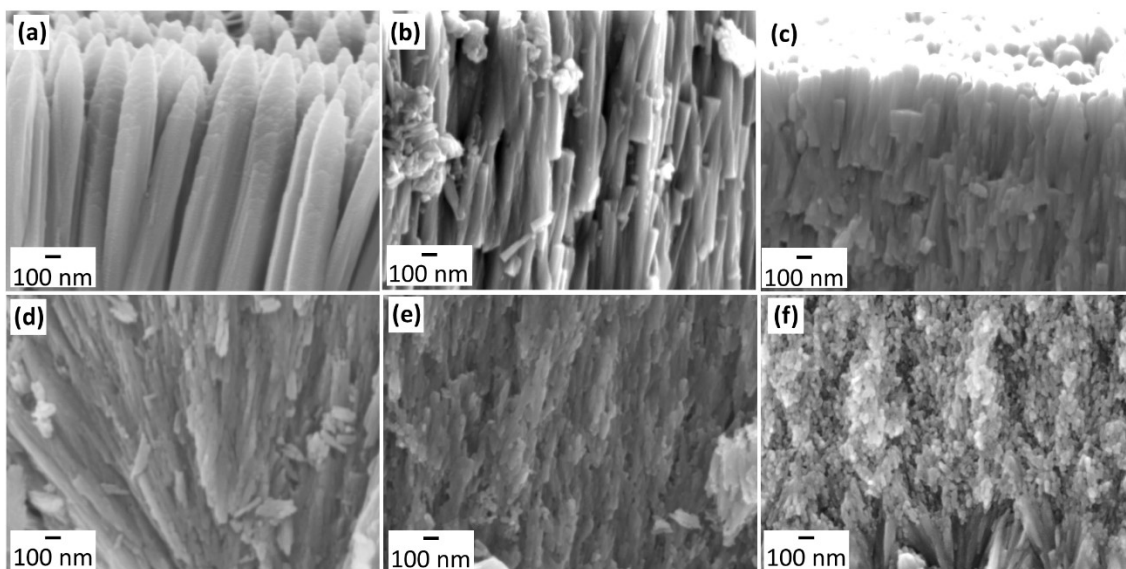


Fig. S6 SEM images (a-f) of the cross-sectional view of the films after subsequent growth at  $[F^-] = 1.50 \text{ mmol dm}^{-3}$  for 1–24 h on the seed layer. (a)  $[Ca^{2+}] = 0.74 \text{ mmol dm}^{-3}$  and  $[PO_4^{3-}] = 0.062 \text{ mmol dm}^{-3}$ , (b)  $[Ca^{2+}] = 0.703 \text{ mmol dm}^{-3}$  and  $[PO_4^{3-}] = 0.279 \text{ mmol dm}^{-3}$ , (c)  $[Ca^{2+}] = 1.361 \text{ mmol dm}^{-3}$  and  $[PO_4^{3-}] = 0.167 \text{ mmol dm}^{-3}$ , (d)  $[Ca^{2+}] = 1.159 \text{ mmol dm}^{-3}$  and  $[PO_4^{3-}] = 0.578 \text{ mmol dm}^{-3}$ , (e)  $[Ca^{2+}] = 2.179 \text{ mmol dm}^{-3}$  and  $[PO_4^{3-}] = 1.085 \text{ mmol dm}^{-3}$ , and (f)  $[Ca^{2+}] = 2.663 \text{ mmol dm}^{-3}$  and  $[PO_4^{3-}] = 1.020 \text{ mmol dm}^{-3}$ . (g) The variation in the width and length of FA nanorods or nanograins with a change in the product of calcium and phosphate ion concentrations.