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## SUPPORTING INFORMATION

## **Controllable Hydrothermal Synthesis of 2D and 3D Dendritic Aluminum Phosphate Crystals**

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**Fig. S1** ED patterns of the dendritic aluminum phosphates prepared at 180 °C for 6 h with molar compositions of (A) 1Al<sub>2</sub>O<sub>3</sub>: 1.3P<sub>2</sub>O<sub>5</sub>: 1.6TEA: 1.3HNO<sub>3</sub>: 425H<sub>2</sub>O and (B) 1Al<sub>2</sub>O<sub>3</sub>: 1.3P<sub>2</sub>O<sub>5</sub>: 1.6TEA: 1.3HNO<sub>3</sub>: 0.05CTAB: 425H<sub>2</sub>O.



<sup>15</sup> **Fig. S2** A low magnification SEM image of dendritic aluminum phosphates prepared at 180 °C for 6 h with molar compositions of 1Al<sub>2</sub>O<sub>3</sub>: 1.3P<sub>2</sub>O<sub>5</sub>: 1.6TEA: 1.3HNO<sub>3</sub>: 425H<sub>2</sub>O.



**Fig. S3** EDX spectra of the 2D dendritic structure in trunk (Spectrum1), branch (Spectrum2) and 3D dendritic structure in trunk (Spectrum3), branch (Spectrum4).



**Fig. S4** TG profiles of the samples prepared at 180 °C for 6 h with molar compositions of (A) 1Al<sub>2</sub>O<sub>3</sub>: 1.3P<sub>2</sub>O<sub>5</sub>: 1.6TEA: 1.3HNO<sub>3</sub>: 425H<sub>2</sub>O and (B) 1Al<sub>2</sub>O<sub>3</sub>: 1.3P<sub>2</sub>O<sub>5</sub>: 1.6TEA: 1.3HNO<sub>3</sub>: 0.05CTAB: 425H<sub>2</sub>O.



<sup>10</sup> **Fig. S5** XRD patterns of the samples prepared at 180 °C for 6 h with HNO<sub>3</sub>/Al<sub>2</sub>O<sub>3</sub> molar ratios of: (a) 0.8, (b) 1.0, (c) 1.6 and (d) 2.0.



**Fig. S6** SEM images of the samples prepared at 180 °C for 6 h with  $HNO_3/Al_2O_3$  molar ratios of (a) 0.8, (b) 1.0, (c) 1.6 and (d) 2.0



**Fig. S7** XRD patterns of the samples prepared at 180 °C for 6 h using (a) HAc, (b) HCl instead of HNO<sub>3</sub> and (c) ammonia, (d) NaOH instead of TEA at the PH value of 3.8, respectively.



**Fig. S8** SEM images of the samples prepared at 180 °C for 6 h using (a) HAc, (b) HCl instead of  $HNO_3$  at the pH value of 3.8, respectively.



s **Fig. S9** XRD patterns of the samples prepared at 180 °C for 6 h with  $P_2O_5/Al_2O_3$  molar ratios of: (a) 1.0, (b) 1.1, (c) 1.2 and (d) 1.4.



**Fig. S10** SEM images of the samples prepared at 180 °C for 6 h with  $P_2O_5/Al_2O_3$  molar ratios of (a) 1.0, (b) 1.1, (c) 1.2 and (d) 1.4.



**Fig. S11** XRD patterns of the samples prepared at 180 °C for 6 h with  $TEA/Al_2O_3$  molar ratios of: (a) 0, (b) 0.4, (c) 0.8, (d) 1.2, (e) 2.0 and (f) 2.5.



**Fig. S12** SEM images of the samples prepared at 180 °C for 6 h with TEA/Al<sub>2</sub>O<sub>3</sub> molar ratios of (a) 0.4, (b) 0.8, (c) 1.2 and (d) 2.0.



Fig. S13 SEM images of the samples prepared at 180 °C for 6 h with  $H_2O/Al_2O_3$  molar ratios of (a) 500, (b) 350.



Fig. S14 XRD patterns of the samples prepared at 180 °C for 6 h with  $CTAB/Al_2O_3$  molar ratios of: (a) 0.10, (b) 0.15, (c) 0.20 and (d) 0.25.



<sup>5</sup> Fig. S15. XRD patterns of the samples synthesized for 6 h at different crystallization temperatures.



**Fig. S16** SEM images of the samples prepared from hydrogels with molar compositions of  $1Al_2O_3$ : 1.3P<sub>2</sub>O<sub>5</sub>: 1.6TEA: 1.3HNO<sub>3</sub>: 425H<sub>2</sub>O at crystallization temperatures of (a) 140 °C, (b) 160 °C and (c) 200 °C for 6 h.