

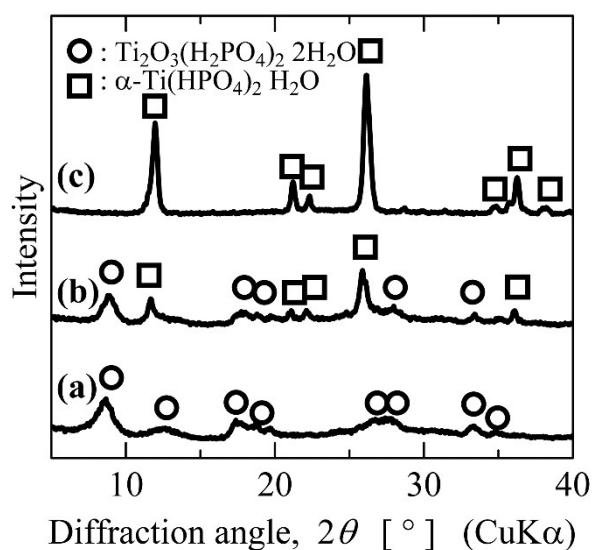
## *Electronic Supplementary Information*

### **Bottom-up synthesis of titanophosphate nanosheets by the aqueous solution process**

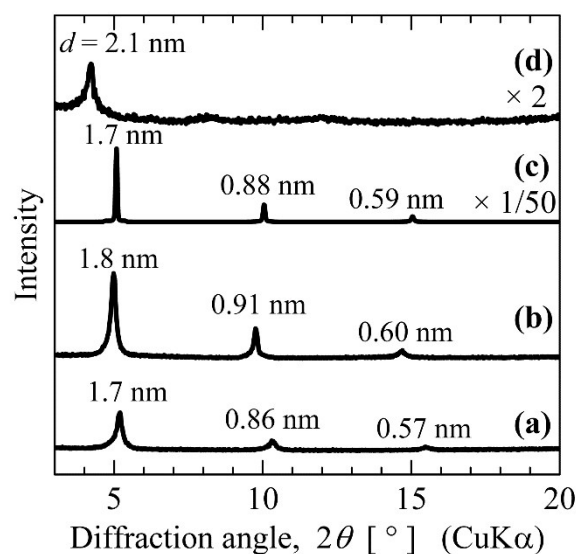
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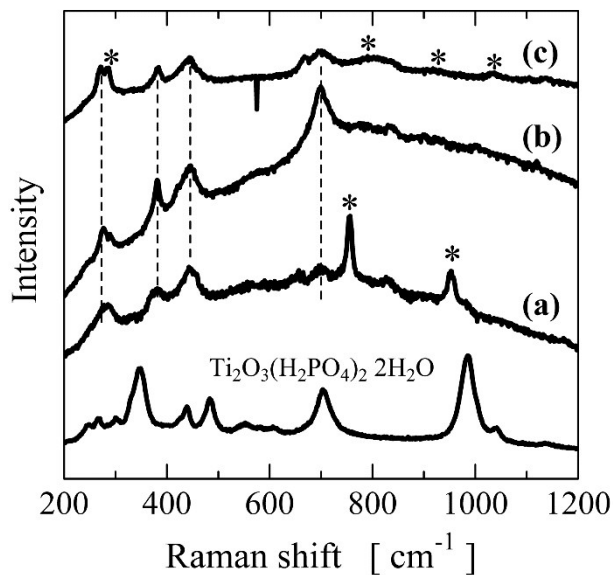
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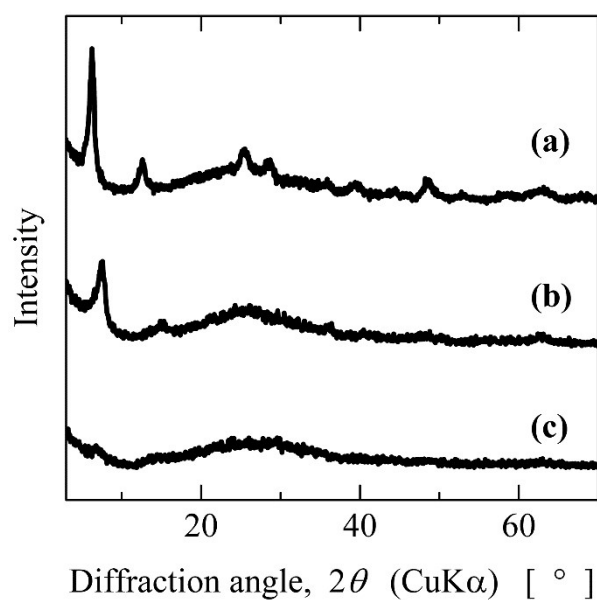
**Figure S1** XRD patterns of the powders prepared by heating the mixtures of TIP and  $\text{H}_3\text{PO}_4$  with  $\text{H}_3\text{PO}_4$  / TIP molar ratios of (a) 1, (b) 10, and (c) 20 at 100  $^{\circ}\text{C}$ .



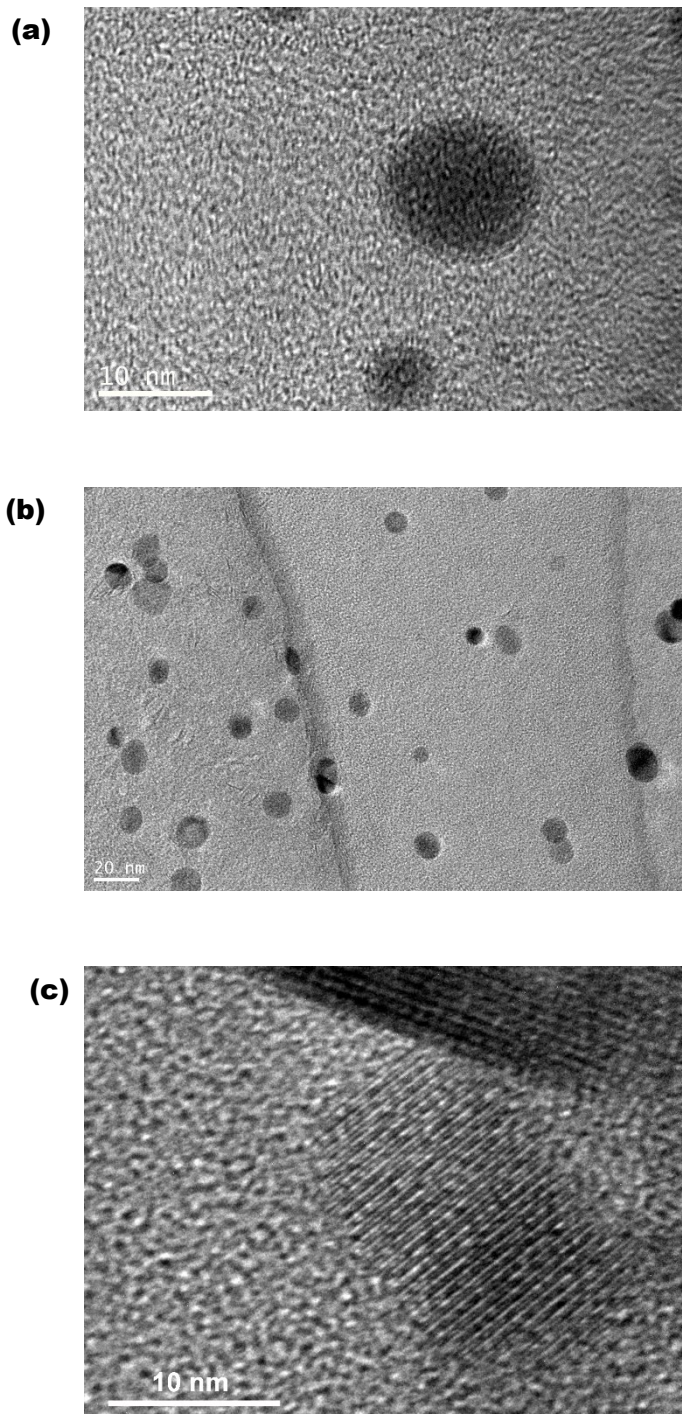
**Figure S2** XRD patterns of the thin films fabricated by evaporating titanophosphate nanosheet sols on a glass substrate. The titanophosphate nanosheet sols were prepared by heating the mixtures of TIP,  $\text{H}_3\text{PO}_4$ , and  $\text{NR}_4\text{OH}$  with a molar ratio of  $\text{TIP} / \text{H}_3\text{PO}_4 / \text{NR}_4\text{OH} = 1 / 1 / 2.2$  at  $80^\circ\text{C}$ . The used  $\text{NR}_4\text{OH}$  was (a) TMAOH, (b) TEAOH, (c) TPAOH, and (d) TBAOH.



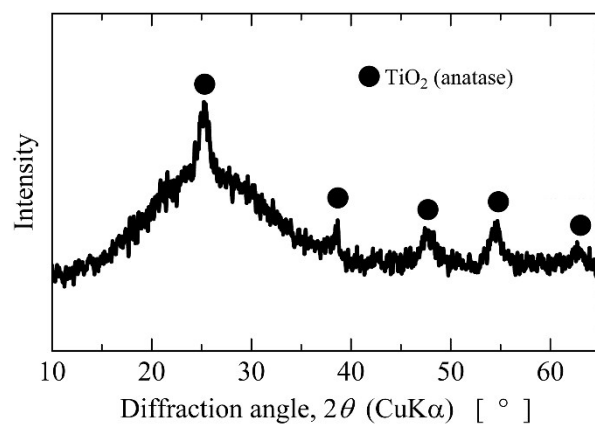
**Figure S3** Raman spectra of the titanophosphate nanosheets prepared by evaporating titanophosphate nanosheet sols and then washed with 2-PrOH. The titanophosphate nanosheet sols were prepared by heating the mixtures of TIP,  $\text{H}_3\text{PO}_4$ , and  $\text{NR}_4\text{OH}$  with a molar ratio of  $\text{TIP} / \text{H}_3\text{PO}_4 / \text{NR}_4\text{OH} =$  (a, b)  $1 / 1 / 2$  and (c)  $1 / 1 / 2.5$  at  $80^\circ\text{C}$ . The used  $\text{NR}_4\text{OH}$  was (a) 2.8 M TMAOH, (b) 1.4 M TEAOH, and (c) 0.5 M TPAOH. The marks \* are assigned to  $\text{NR}_4^+$  ions. For comparison, the Raman spectrum of layered titanium phosphate  $\text{Ti}_2\text{O}_3(\text{H}_2\text{PO}_4)_2 \cdot 2\text{H}_2\text{O}$  is also shown.



**Figure S4** XRD patterns of the powders prepared from the titanophosphate nanosheet sols synthesized from the sols (a) prepared at TIP / H<sub>3</sub>PO<sub>4</sub> / TPAOH = 1 / 1 / 2.5 using 0.5 M TPAOH and prepared at TIP / H<sub>3</sub>PO<sub>4</sub> / TEAOH = 1 / 1 / (b) 2.0 and (c) 2.2 using 1.4 M TEAOH. The sample (a), (b), and (c) correspond to O (large),  $\Delta$  (medium), and  $\times$  (small), respectively.



**Figure S5** TEM images of the titanophosphate nanosheets synthesized (a) from the aqueous mixture with a molar ratio of  $\text{TIP} / \text{H}_3\text{PO}_4 / \text{TPAOH} = 1 / 1 / 2.2$  in very weakly basic aqueous sols with a pH value of about 9, (b) from the aqueous mixture with a molar ratio of  $\text{TIP} / \text{H}_3\text{PO}_4 / \text{TEAOH} = 1 / 1 / 2$ , which was prepared by using 1.4 M TEAOH solution, and (c) from the aqueous mixture with a molar ratio of  $\text{TIP} / \text{H}_3\text{PO}_4 / \text{TBAOH} = 1 / 1 / 2$ , which was prepared by using 1.5 M TBAOH solution.



**Figure S6** XRD pattern of the precipitates formed by adding titanophosphate nanosheet powder including an amorphous phase to copper acetate aqueous solution.