**Electronic Supplementary Information**

**Bottom-up synthesis of titanate nanoflakes with euhedral shapes by aqueous solution process**

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**Figure S1**  TEM image of closely packed titanate nanoflakes with a euhedral shape, which is a rhombic shape. They are oriented along three different crystallographic directions. They were synthesized according to our previous study, as follows: They were obtained by heating aqueous mixture with a molar ratio of TIP/ TBAOH/ teaH$_3$ = 1: 0.5: 2 and a Ti concentration of 0.5 M at 80 °C for 7 days, followed by dialyzing the resulting sol for 1 h.
Figure S2  TEM image of the titanate nanoflakes synthesized by heating a reaction sol at 80 °C for 1 week. The reaction sol had a Ti concentration of 0.1 M and a mixing molar ratio of TIP/ TBAOH = 1: 0.5.
**Figure S3** Crystallization profiles of titanate nanoflakes under different synthetic conditions. XRD measurements were made for the thin films fabricated by evaporating the heated sols. The amount of formed titanate nanoflakes were evaluated from the intensity of the (020) peak of layered titanates, which were formed by stacking up titanate nanoflakes. (a) The reaction sols with a TIP/ TMAOH molar ratio of 1: 0.5 and different Ti concentrations were heated at 80 °C. (b) The reaction sols with a TIP/ TMAOH molar ratio of 1: 0.5 and a Ti concentration of 100 mM were heated at 80 °C or 100 °C. (c) The reaction sols with a Ti concentration of 100 mM and different TIP/ TMAOH molar ratios were heated at 80 °C.
Figure S4  TEM images of the titanate nanoflakes synthesized from the aqueous mixture with a molar ratio of TIP / TMAOH = 1: 0.5 and a Ti concentration of 0.1 M by heating 80 °C for 1 week. Despite the fact that the titanate nanoflakes surrounded with broken lines had indistinct shapes, they aggregated by closely packing with a high orientation.

Figure S5  TEM image and selected-area electron diffraction of the titanate nanoflake synthesized from the aqueous mixture with a molar ratio of TIP/ TMAOH/ H\textsubscript{2}lac = 1: 2.5: 2 and a Ti concentration of 20 mM by heating 60 °C for 3 weeks. The titanate nanoflake was surrounded with the \{100\} and \{101\} planes.
**Figure S6** XRD patterns of the thin film fabricated by drying titanate nanoflake sol on a glass substrate, the powder prepared by evaporating the sol with a rotary evaporator, and the wet powder prepared by evaporating the sol and then wetting the resulting powder with several water drops. The titanate nanoflake sol was prepared by mixing TIP, TBAOH, and H\textsubscript{3}tea in water with a molar ratio of TIP/ TBAOH / H\textsubscript{3}tea = 1: 0.5: 3 and a Ti concentration of 0.1 M and then heating resulting aqueous mixture at 80 °C for 7 days. Moreover, the interplanar spacing \((d)\) and the plane index \((hkl)\) of the lattice plane providing a diffraction peak are shown on corresponding peaks.
Figure S7  XRD patterns of the wet powders by evaporating titanate nanoflake sols with a rotary evaporator, placing resulting powders on XRD sample holders, and then wetting the powders with several water drops. The titanate nanoflake sols were prepared by mixing TIP with TMAOH or TBAOH in water with or without the addition of H$_3$tea and then heating resulting aqueous mixture at 80 °C. The heating period was 7 days for TIP/ TBAOH/ H$_3$tea sample and 1 day for the others. Moreover, the interplanar spacing ($d$) and the plane index ($hkl$) of the lattice plane providing a diffraction peak are shown on corresponding peaks.
Figure S8  TEM image of the titanate nanoflakes synthesized by heating an aqueous mixture of TIP, H$_3$tea, and TBAOH at 80 °C for 7 days. This figure is the same as Figure 7b. For clearly showing the formation of titanate nanoflakes with a euhedral shape, some nanoflakes were surrounded with broken lines.

Figure S9  TEM images of the titanate nanoflakes obtained by further growing the preformed nanoflakes in a fresh reaction sol. The reaction sol was prepared with a molar ratio of TIP/ H$_3$tea/ TBAOH = 1/ 3/ 0.5 and a Ti concentration of 0.1 M. The preformed titanate nanoflakes were synthesized by heating the reaction sol at 80 °C for 1 week. The obtained sol was mixed with a fresh reaction sol at volume ratios of (a) 1:7 and (b) 1: 3. Then, the mixture was heated again at 80 °C.
Figure S10  (a) TEM image of the titanate nanoflakes synthesized from the aqueous mixture with a Ti concentration of 0.5 M and a molar ratio of TIP/ TBAOH / H$_3$tea = 1: 0.5: 3 by heating 80 °C for 1 week.\textsuperscript{S1}  
(b) TEM image of the titanate nanoflakes obtained by dialyzing the ones shown in (a) for 1 hour in order to remove the triethanolamine molecules adsorbed on the edge of nanoflakes.\textsuperscript{S1} The nanoflakes shown in (b) is the same as the ones shown in Figure S1.

Reference