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Synthesis and Growth Mechanism of Multilayer TiO₂ Nanotube Arrays

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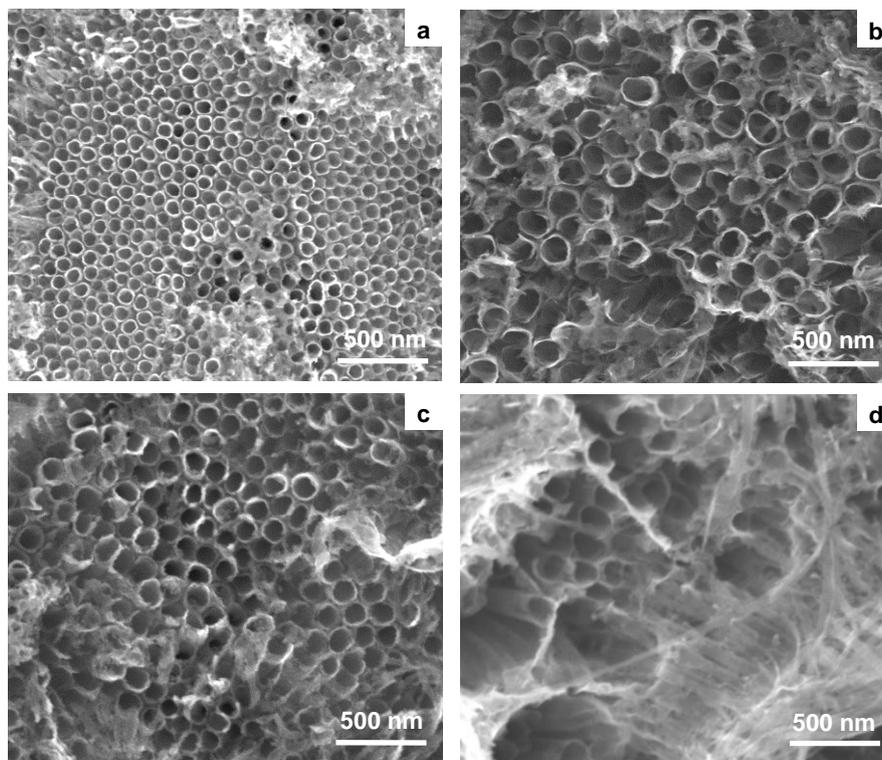


Figure S1. SEM images showing top-view of various TiO₂ nanotube arrays formed in EG electrolytes containing 0.3 wt% NH₄F and 5 vol% H₂O via different anodization sequences: (a) 60 V for 10 min, followed by 10 V for 10 min ($t_2/t_1 = 1:1$) and 60 V for 10 min (short double-layer nanotubes); (b) 60 V for 2 h (single-layer nanotubes), (c) 60 V for 60 min, followed by 10 V for 10 min ($t_2/t_1 = 1:6$) and 60 V for 60 min (bamboo-type nanotubes); (d) 60 V for 60 min followed by 10 V for 60 min ($t_2/t_1 = 1:1$) and 60 V for 60 min (double-layer nanotubes).

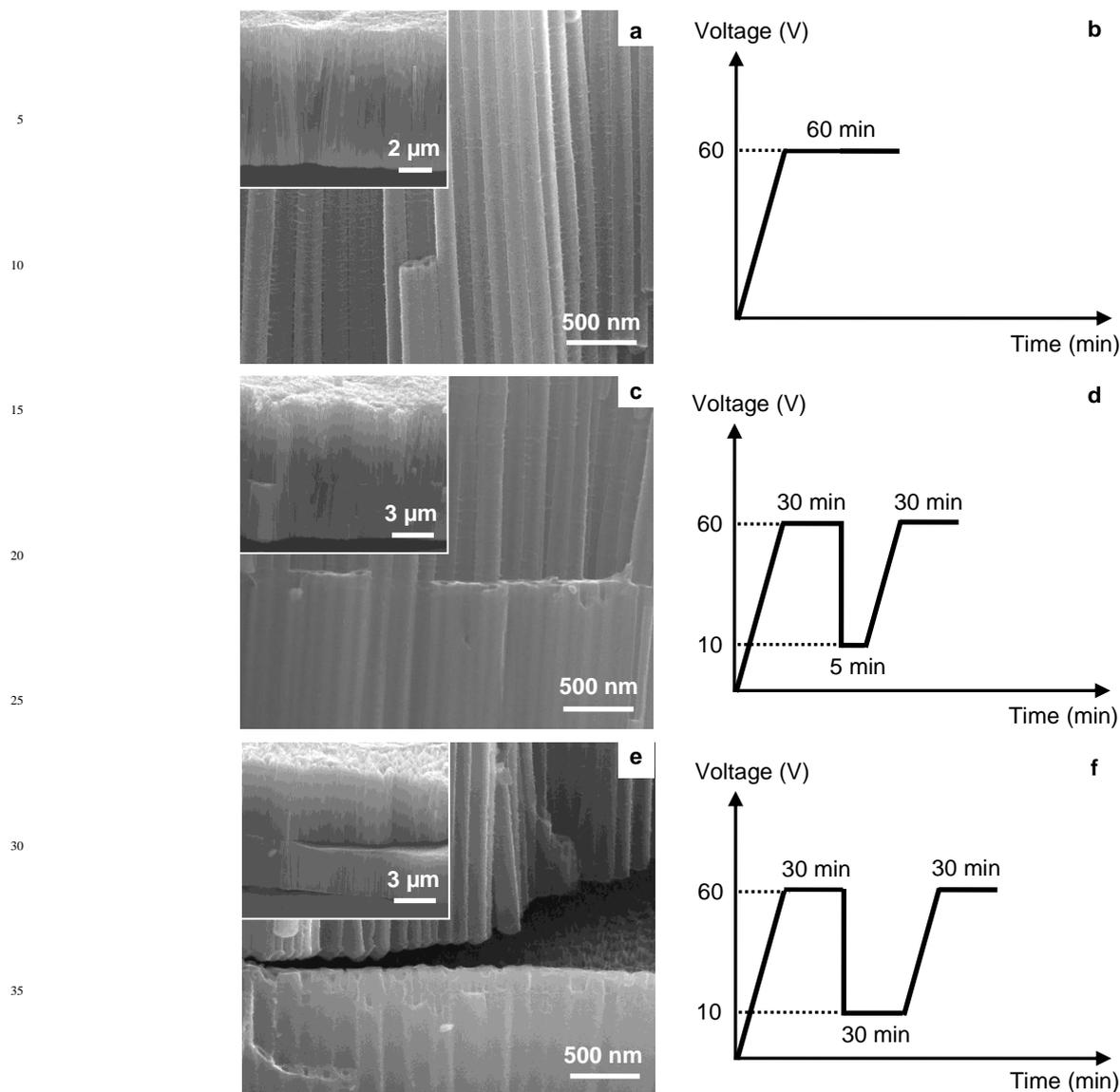
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40 **Figure S2.** Cross-section SEM images and corresponding anodization sequences of TiO₂ nanotube arrays formed in EG electrolytes containing 0.3 wt% NH₄F and 5 vol% H₂O: (a-b) 60 V for 60 min; (c-d) 60 V for 30 min followed by 10 V for 5 min ($t_2/t_1 = 1:6$) and 60 V for 30 min; (e-f) 60 V for 30 min followed by 10 V for 30 min ($t_2/t_1 = 1:1$) and 60 V for 30 min.

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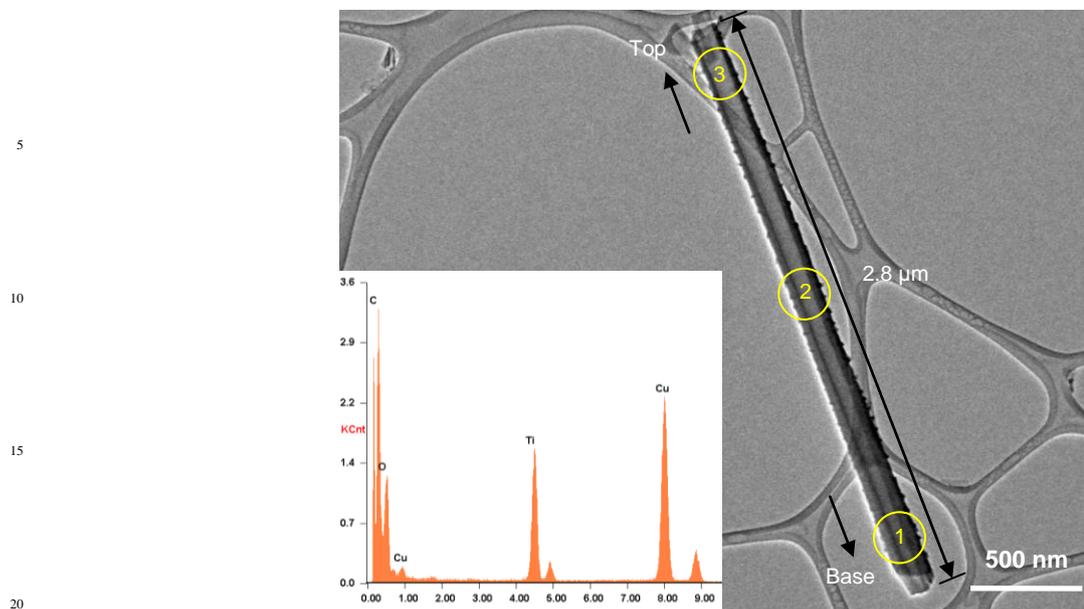


Figure S3. TEM image and EDS spectra (inset) of a nanotube in single-layer TiO₂ nanotube arrays formed at 60 V for 2 h in EG electrolyte containing 0.3 wt% NH₄F and 5 vol% H₂O (Note: EDS analysis was performed at spots 1, 2 and 3, respectively).

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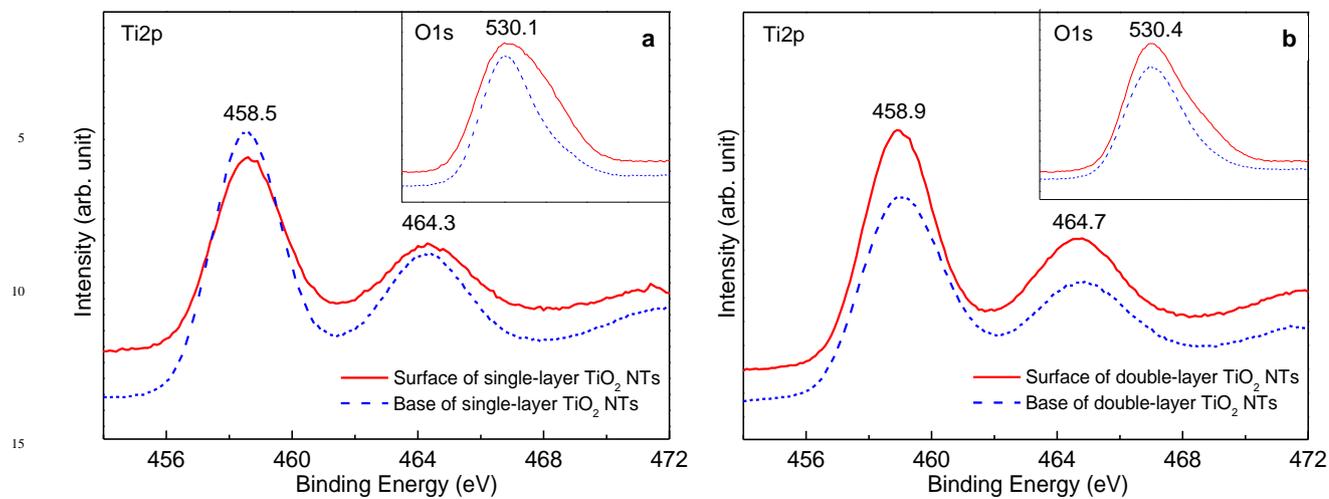


Figure S4. Ti2p XPS peaks and O1s XPS peaks (insets) taken from the top and the bottom of (a) single-layer TiO₂ nanotubes synthesized at 60 V for 2 h, and (b) double-layer TiO₂ nanotube arrays synthesized at 60 V for 60 min followed by 10 V for 60 min ($t_2/t_1 = 1:1$) and 60 V for 60 min, in EG electrolytes containing 0.3 wt% NH₄F and 5 vol% H₂O.

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