[Supporting Information]

Facile and Accelerated Production of RuO₂ Monolayers via a Dual-Step Intercalation Process

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S1. Thickness of exfoliated RuO₂ monolayer nanosheets



Figure S1. Atomic force microscopy (AFM) image of exfoliated RuO_2 monolayer nanosheets. The average thickness of the RuO_2 monolayer nanosheets is 0.86 nm.

S2. Structure information of exfoliated RuO₂ monolayer nanosheets



Figure S2. (a) Selected area diffraction pattern (SADP) of an exfoliated RuO₂ monolayer nanosheet. (b) Atomic structures of the RuO₂ nanosheet with the definitions of the lattice parameters (a, b, θ , ϕ).

S3. The yield and concentration of RuO₂ monolayers derived from the UV-vis spectroscopy

The concentration of RuO_2 monolayers was proportional to the absorbance at 360 nm (ref: Journal of Solid State Chemistry 182, 2997 (2009)), and the absorbance was measured by UVvis spectroscopy. The equation of the RuO_2 concentration is as below.

$$C = \frac{AM_w}{\alpha l} \tag{eq 1}$$

C is concentration of the RuO₂ monolayers, A is absorbance at 360 nm, M_w is molar weight of RuO₂, l is the length of cuvette, and α is the molar extinction coefficient of the dispersed RuO₂ monolayers in aqueous solution, which is 7.4 x 10³ mol⁻¹dm³cm⁻¹ at 360 nm (ref: Journal of Solid State Chemistry 182, 2997 (2009)).

The yield of the RuO_2 monolayers was obtained by dividing the concentration of RuO_2 monolayers to initial RuO_2 concentration. The equation of the yield of the RuO_2 concentration is as below.

$$Y = \frac{AM_w L}{\alpha lm} \tag{eq 1}$$

Y is the yield of the RuO_2 monolayers, L is an initial volume of aqueous solution, and m is an initial weight of H_xRuO_2 layered materials.

Figure S1a shows UV-vis absorption spectra of the aqueous solution containing RuO_2 monolayers with TBA⁺ as an intercalant. Also, Figure S2a shows UV-vis absorption spectra of the RuO_2 monolayer solution using TBA⁺ and TMA⁺ as intercalants.



Figure S3. UV-vis absorption spectra of the aqueous solution containing RuO_2 monolayers using (a) TBA⁺ as an intercalant, and (b) TBA⁺ and TMA⁺ as intercalants.

S4. Atomic structures of DFT (Density Functional Theory) simulation



Figure S4. Atomic structures of DFT (Density Functional Theory) simulation. (a) RuO_2 layered structure, (b) TMA⁺ intercalated, and (c) TBA⁺ intercalated RuO_2 layered structures. (d) RuO_2 layered structure without strain. (e) 2 layers in the middle of the RuO_2 layered structure are widened from 0.51 to 0.92 nm for intercalation of TMA⁺. (f) The distance between the RuO_2 layers are pulled apart from 0.51 to 1.24 nm for intercalation of TBA⁺. (g) TMA⁺ intercalated RuO_2 layered structure without strain. (h) The RuO_2 layers in the middle of the structure are widened from 0.92 to 1.24 nm for intercalation of TBA⁺. Red, green, grey, blue and white balls represent oxygen, ruthenium, carbon, nitrogen, and hydrogen atoms, respectively.