

# Supporting Material

## **Preparation of aqueous zinc ion rGH/V<sub>2</sub>O<sub>5</sub> photorechargeable supercapacitor**

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## 2 Experimental

### 2.1 Preparation of GO and rGH

The preparation process diagram of GO and rGH was shown in Fig. 1(b), and the detailed preparation process was as follows. Initially, 1.5 g of MG, 1.5 g of NaNO<sub>3</sub> and 48 mL H<sub>2</sub>SO<sub>4</sub> were added in a beaker, and then 9 g of KMnO<sub>4</sub> was slowly added to a beaker under stirring condition. Subsequently, the beaker was placed in a oil bath at 30 °C with stirring for 15 h. Then, 85 mL of deionized water was added to the above mixed liquid, and the temperature of the oil bath was raised to 80 °C, and kept for 35 min. Finally, 100 mL of deionized water was added to the beaker with stirring condition, and 8 mL of H<sub>2</sub>O<sub>2</sub> was added until the suspension turned to bright yellow. The bright yellow suspension was washed using HCl (5 %) solution and deionized water several times until the pH of the filtrate was equal to 7, so GO suspension was obtained. Finally, the appropriate amount of GO suspension was placed in an autoclave at 180 °C for 4 h to prepare rGH.

## 3 Results and discussion

### 3.1 Photoelectric performance of the rGH/V<sub>2</sub>O<sub>5</sub> supercapacitor

The specific capacities of the supercapacitors with different mass ratios of V<sub>2</sub>O<sub>5</sub> to rGH are shown in Table 1S. With mass ratios ranging from 1:1 to 3:1, the specific capacities first increase and then decrease. When the mass ratio of V<sub>2</sub>O<sub>5</sub> to rGH is 2:1, the supercapacitor has the highest specific capacity, and the values are 164 F. g<sup>-1</sup> and 136.1 F. g<sup>-1</sup> without and with illumination, respectively.

Because the mass ratios of V<sub>2</sub>O<sub>5</sub> to rGH are too small, the photoanode films are relatively thinner. For example, the mass ratio of V<sub>2</sub>O<sub>5</sub> to rGH is 1:1, the thickness of the photoanode film is approximately 1.0 μm, which leads to lower light absorption and photoelectric performance.<sup>1</sup> When the mass ratios are too large, the photoanode films are relatively thicker. For example, the mass ratio of V<sub>2</sub>O<sub>5</sub> to rGH is 3:1, the thickness of the photoanode film is approximately 3 μm, which results in carrier recombination,<sup>2</sup> thereby reducing the specific capacity. When the mass ratio of V<sub>2</sub>O<sub>5</sub>: rGH is 2 :1, the photoanode film has an appropriate thickness of approximately 2 μm. It can sufficiently harvest light energy, converting it into electric energy,<sup>3</sup> thereby minimizing carrier

recombination and consequently enhancing the specific capacity.

**Table 1S** Effect of mass ratios of V<sub>2</sub>O<sub>5</sub> to rGH in the electrodes on the specific capacities of the supercapacitors (with a current density of 0.5 A. g<sup>-1</sup>)

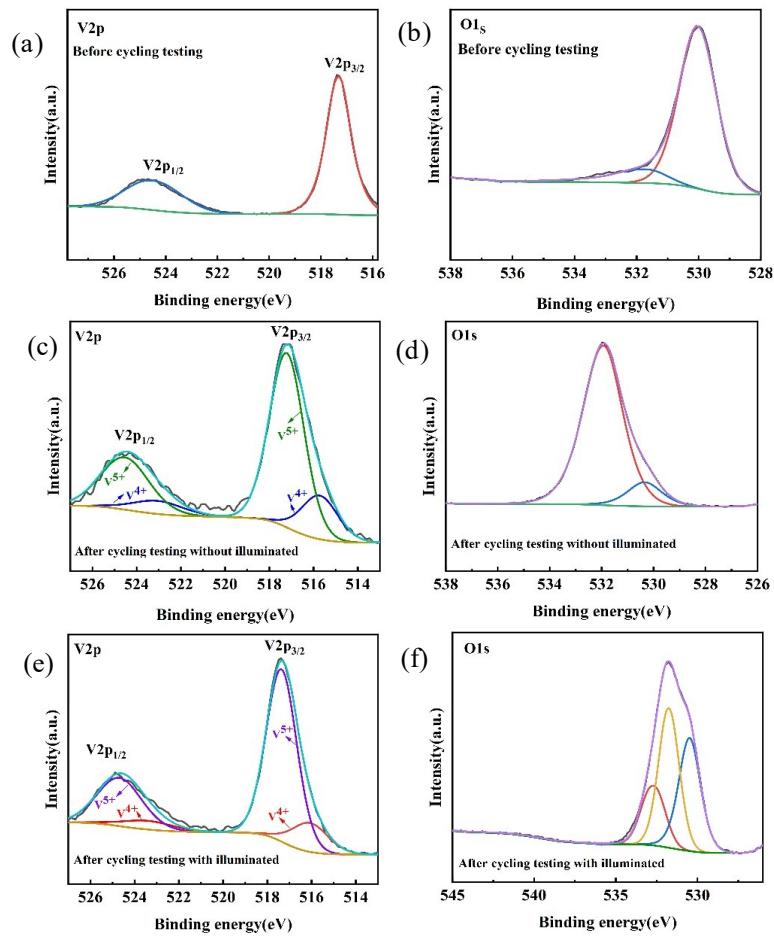
V <sub>2</sub> O <sub>5</sub> : rGH	1:1	1.5:1	2:1	2.5:1	3:1
Without illumination	77.3	92.1	136.1	109.1	86.2
With illumination	91.1	113	164	142.4	101.2

Table 2S shows photoelectrochemical performance of the supercapacitors about similar literature and this work. It can be seen that the synthesized device has a higher photocharging voltage (0.82 V), the specific capacity (25.25F. g<sup>-1</sup>) and PCE (0.012%) compared with similar literature, indicating that the synthesized device has perfect comprehension performance.

**Table 2S** Comparison of photoelectrochemical performance between similar literature and our work

Photo-rechargeable energy storage unit	Photo-charge conversion efficiency	The specific capacity	Photovoltage	Energy density	Refs
TiO <sub>2</sub> / dye/ LiI/ AC based photo-rechargeable supercapacitors	-	0.69 F cm <sup>-2</sup>	0.45 V	-	[4]
3D Hybrid Halide perovskite photo-rechargeable supercapacitors	0.02% @ 20 mW cm <sup>-2</sup>	-	0.325 V	0.16 Wh kg <sup>-1</sup>	[5]
ITO BR/P3HT based photo-rechargeable supercapacitors	0.0017% @ 100 mW cm <sup>-2</sup>	2.44 mF cm <sup>-2</sup>	0.28 V	-	[6]
2D(PEA) <sub>2</sub> PbBr <sub>4</sub> / 3D MAPbBr <sub>3</sub> based photo-rechargeable supercapacitors	0.03% @ 30 mW cm <sup>-2</sup>	-	0.119 V	6.4 W h kg <sup>-1</sup>	[7]
g-C <sub>3</sub> N <sub>4</sub> /Zn-ion capacitors	0.01% @ 420nm illumination	11.377 F g <sup>-1</sup>	0.85 V	0.7 Wh kg <sup>-1</sup>	[8]
rGH/V <sub>2</sub> O <sub>5</sub>	0.012% @ 95 mW cm <sup>-2</sup>	25.25 F g <sup>-1</sup>	0.82 V	2.36 Wh kg <sup>-1</sup>	This work

Fig.1S shows XPS spectra of the  $V_2O_5$  photoanodes before and after 10000 cycles. As shown in Fig.1S (a-b), the  $V2p_{3/2}$  peak at 517.3 eV and the  $V2p_{1/2}$  peak at 524.45 eV correspond to  $V^{5+}$  characteristic binding energies<sup>9</sup>, and the two peaks of  $O1s$  at 530.2 eV and 531.8 eV correspond to V-O bond and C-O bond. After 10000 cycles, it can be seen from Fig.1S (c) and (e), there is a little of amount of  $V^{4+}$  peak due to the redox reaction during cycling, and the peak area of  $O1s$  at 531.8eV (see Fig.1S(d) and(f)) has a little increase due to the presence of  $V^{4+}$ . However, there is no significant change in the structure of  $V_2O_5$  before and after cycling.



**Fig.1S** XPS spectra of the  $V_2O_5$  photoelectrode (a-b) before, and (c-d) without illuminated, (e-f) with illuminated after 10000 cycles.

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