## **Electronic Supplementary Information**

## Bifunctional NiCuO<sub>x</sub> photoelectrodes to promote pseudocapacitive charge storage by *in-situ* photocharging

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Fig. S1 Calculated work functions (a and b) of NiO (110) and CuO (010), and charge density distribution contour (c) of the NiO-CuO interface.



Fig. S2 EDS results of the sample CC@NiCuO<sub>x</sub>.



Fig. S3 SEM images of the CC@CuO<sub>x</sub>.



Fig. S4 SEM images of the CC@NiO.







Fig. S6 Valence band curves obtained from the UPS spectra of the CC@NiO (a),  $CC@CuO_x$  (b), and  $CC@NiCuO_x$  (c) electrodes.



Fig. S7 Comparison of ESR results for the three samples.



Fig. S8 CV curves obtained from various scan rates (a) with a potential window of 0-0.6 V (vs. SCE) and GCCD curves obtained from various current densities (b) of the  $CC@NiCuO_x$  electrodes without light irradiation.



Fig. S9 Comparison of CV curves of CC@NiCuO<sub>x</sub> electrodes with light on/off conditions performed at specific scan rates of 8 mV s<sup>-1</sup> (a), 10 mV s<sup>-1</sup> (b), 15 mV s<sup>-1</sup> (c), 20 mV s<sup>-1</sup> (d), 30 mV s<sup>-1</sup> (e), and 50 mV s<sup>-1</sup> (f), respectively.



Fig. S10 Comparison of GCCD curves of CC@NiCuO<sub>x</sub> electrodes with light on/off conditions performed at specific current densities of 5 mA cm<sup>-2</sup> (a), 8 mA cm<sup>-2</sup> (b), 15 mA cm<sup>-2</sup> (c), 20 mA cm<sup>-2</sup> (d), 30 mA cm<sup>-2</sup> (e), and 50 mA cm<sup>-2</sup> (f), respectively.



Fig. S11 CV curves (a-b) obtained from various scan rates with a potential window of 0-0.6 V (vs. SCE) without (a) and with (b) light irradiation, and GCCD curves (c-d) obtained from various current densities without (c) and with (d) light irradiation of the CC@CuO<sub>x</sub>.



Fig. S12 CV curves (a-b) obtained from various scan rates with a potential window of 0-0.6 V (vs. SCE) without (a) and with (b) light irradiation, and GCCD curves (c-d) obtained from various current densities without (c) and with (d) light irradiation of the CC@NiO.



Fig. S13 Comparison of capacitance enhancements calculated at various current densities of the  $CC@CuO_x$  with light on and off conditions.



Fig. S14 Comparison of capacitance enhancements calculated at various current densities of the CC@NiO with light on and off conditions.

Photoelectrodes	Specific Capacitance	Stability	References
CC@NiCuO <sub>x</sub>	2937 mF cm <sup>-2</sup> at 5 mA cm <sup>-2</sup>	~100% after 5000 cycles	This work
TiO <sub>2</sub> /CNT fibers	$0.6 \text{ mF cm}^{-2} \text{ at } 0.25 \ \mu\text{A}$	/	1
TiO <sub>2</sub> nanotube arrays	1.1 mF cm <sup>-2</sup> at 1 mA cm <sup>-2</sup>	98.8% after 3000 cycles	2
Bi-polar TiO <sub>2</sub> nanotube arrays	262.5 mF cm <sup>-2</sup> at 1 mA cm <sup>-2</sup>	~90% after 5000 cycles	3
TiO <sub>2</sub> /SNGP/CdS	104.6 mF cm <sup>-2</sup> at 1 mA cm <sup>-2</sup>	~85% after 5000 cycles	4
CF/TiO <sub>2</sub> /MoS <sub>2</sub> fibers	18 mF cm <sup>-2</sup> (1740 μF cm <sup>-1</sup> ) at 0.02 mA	~81% after 1000 cycles	5
PANI/CNT composites	422 mF cm <sup>-2</sup> at 0.02 mA	~96% after 500 cycles	6
3D porous graphene and polypyrrole	2754 mF cm <sup>-2</sup> at 0.5 mA cm <sup>-2</sup>	85.8% after 10000 cycles	7
Indium tin oxide NWs	2.44 mF cm <sup>-2</sup> at 0.02 mA cm <sup>-2</sup>	93% after 10000 cycles	8

Table S1. Comparison of the as-prepared CC@NiCuO<sub>x</sub> bifunctional materials with other photoelectrodes for supercapacitors.

## References

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