Supplementary File

Zinc oxide nanoflake/reduced graphene oxide nanocomposite-based dual-acting electrodes for solar-assisted supercapacitor applications

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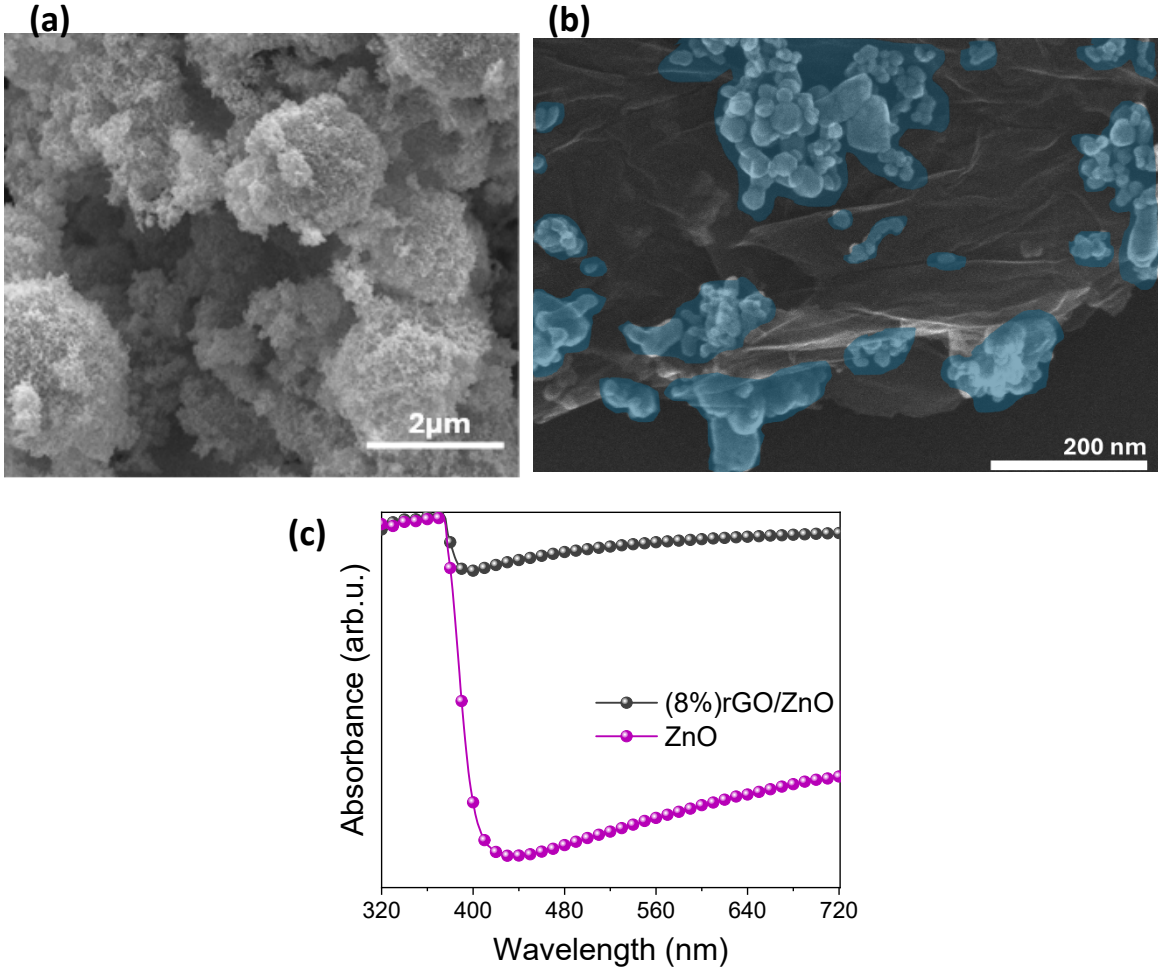


Fig. S1. (a) High magnification SEM image of ZnO nanoflake, (b) SEM image of (16%)rGO/ZnO composite (Blue colored areas represent ZnO), (c) Comparative UV-VIS absorption curves of the pristine ZnO nanoflakes and the (8%)rGO/3D ZnO composite.

Table S1. PL peaks deconvolution emission percentages.

Emissions	(32%)rGO/3D ZnO (%)	(16%)rGO/3D ZnO (%)	(8%)rGO/3DZnO (%)
UV	53	36	38
Blue	26	43	43
Green	11	6	6
Yellow	10	15	13

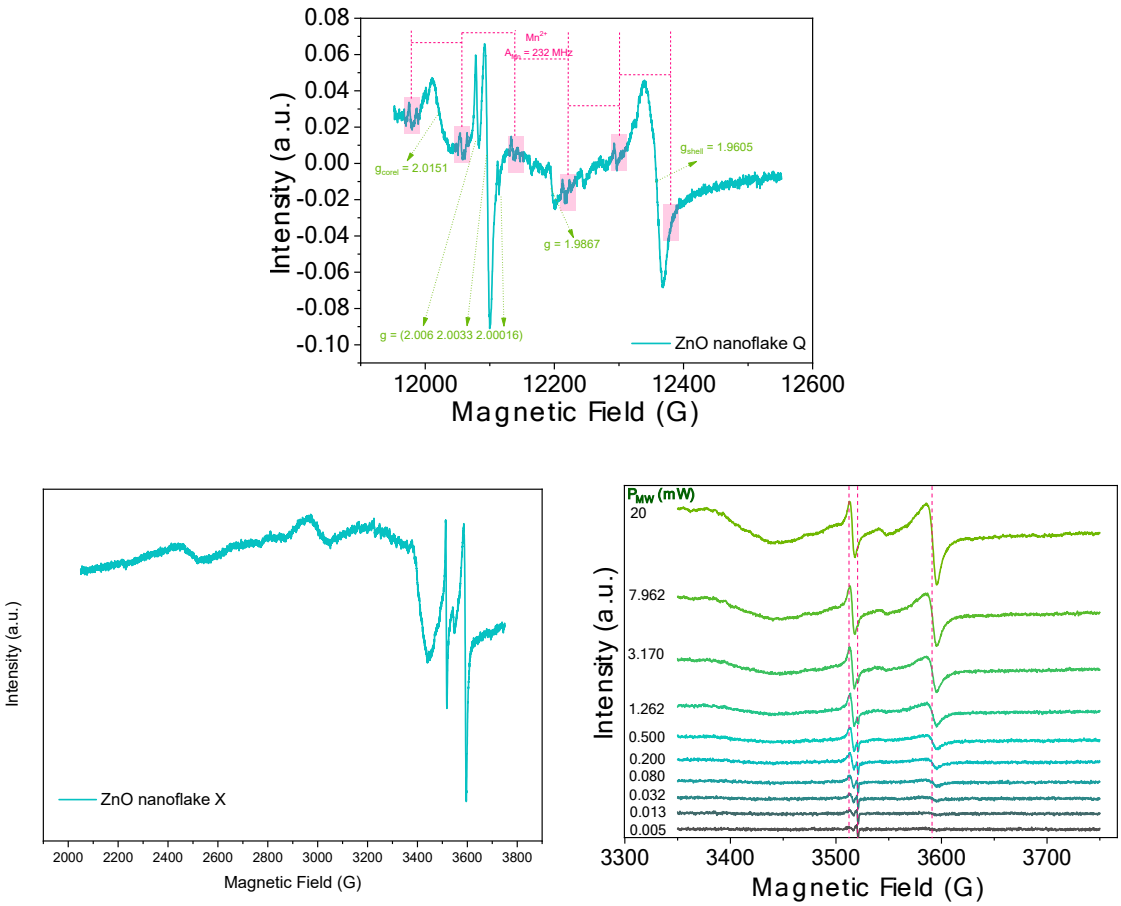
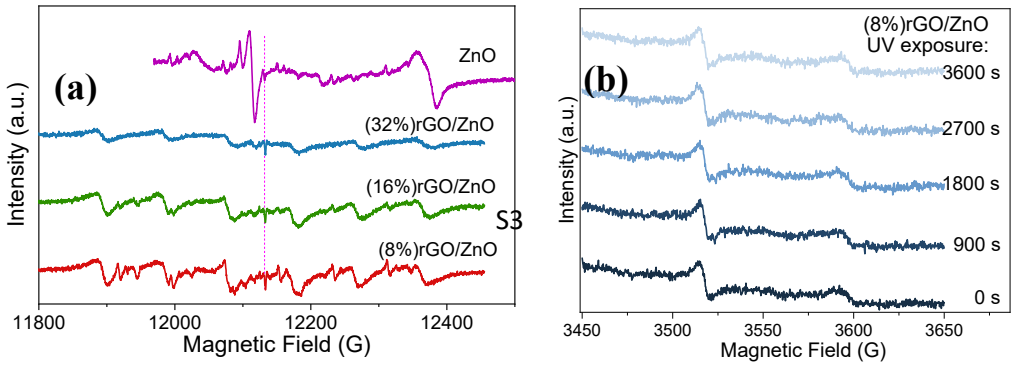


Fig. S2. EPR analysis results for the pristine 3D ZnO nanoflake; (a) at the Q-band, (b) at the X-band, (c) change in signals depending on power intensity.



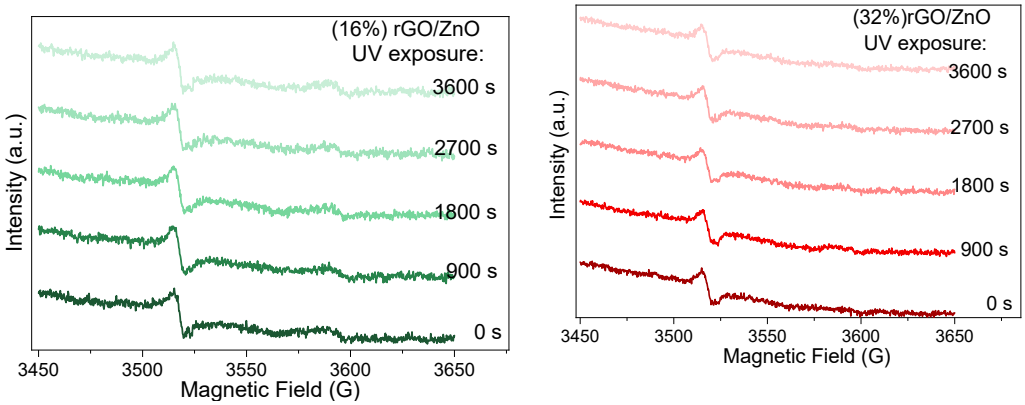


Fig. S3. EPR analysis results for the composite powders

Equations for (Photo)electrochemical measurements

The areal capacitance (C_A) of the PSC devices was estimated from the CV curves using Equation (1)[1]

$$C_{A,CV} = \frac{I\int dV}{AxvxΔV^2} \quad (1)$$

where $\int I\, dV$ is the area under the curve calculated from CV, v is the scan rate, ΔV is the potential window, and A is the active area of the electrode.

C_A can be calculated from GCD curves using the following equation,

$$C_{A,GCD} = \frac{IxdV}{Axdt} \quad (2)$$

Herein, I is the applied current, A is the active area of the electrode, dt is the time for discharging process and V is the potential window. The areal energy density (E_A, μWhcm^{-2}) and power density (P_A, mWcm^{-2}) of the electrodes were estimated using the equation (3) and (4), respectively.[2]

$$E_A = \frac{0.5xC_AxV^2}{3600} \quad (3)$$

$$P_A = \frac{E_Ax3600}{\Delta t} \quad (4)$$

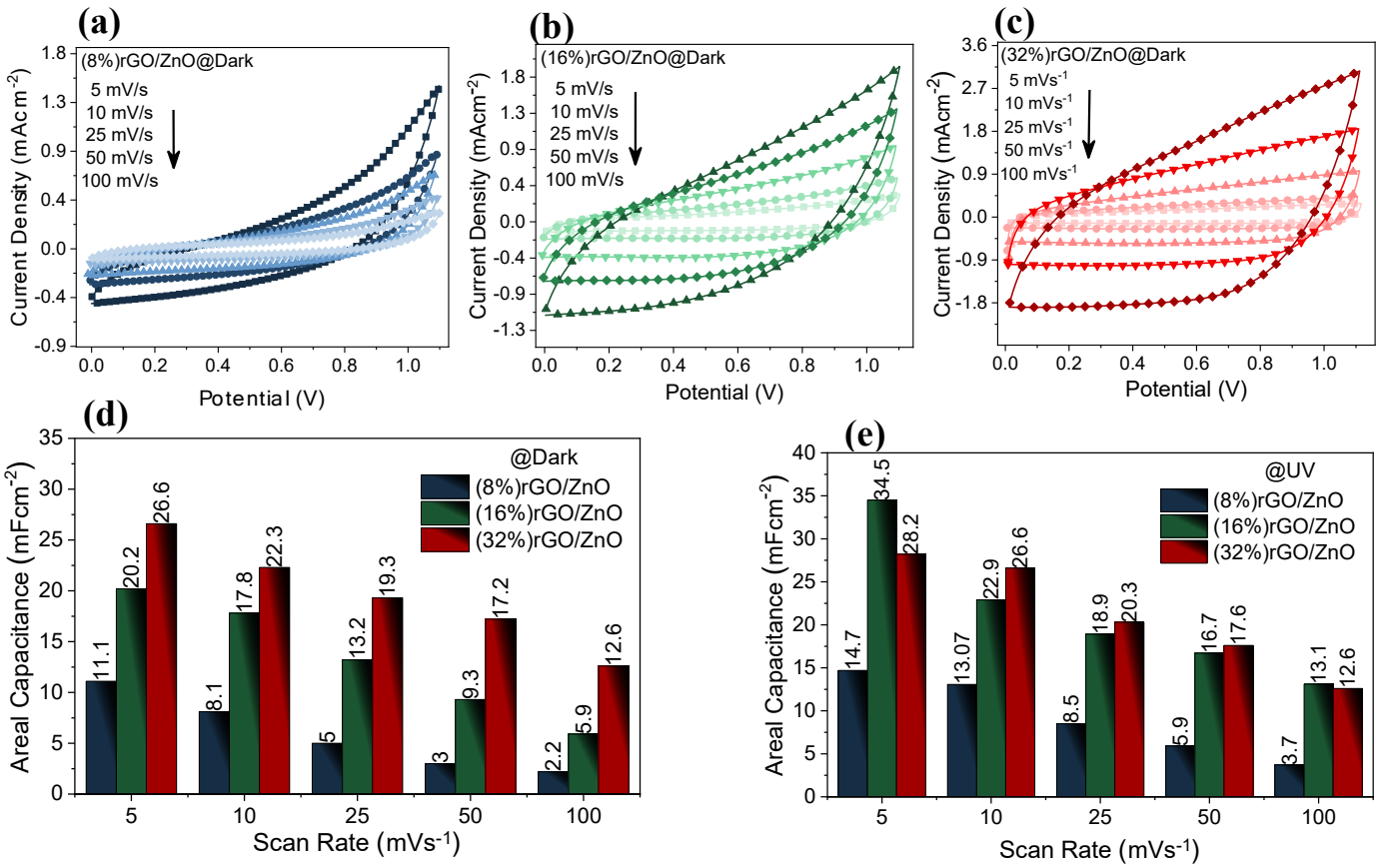
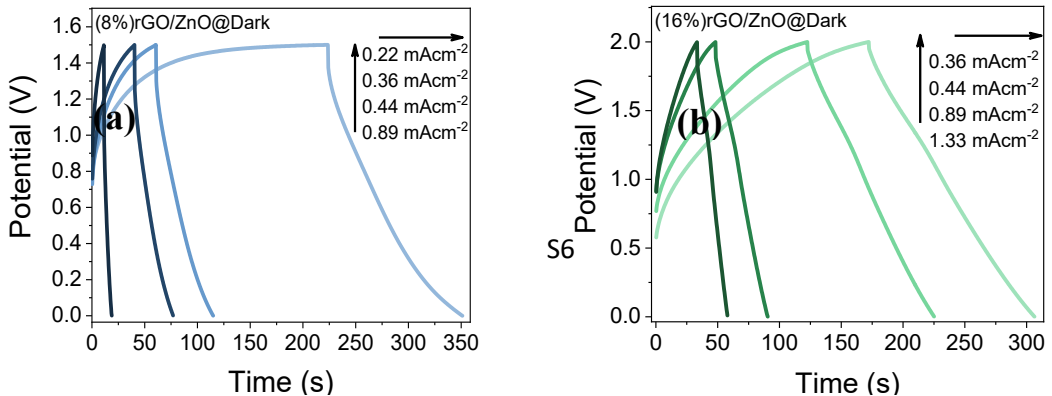


Fig. S4. Dark CV scans at various scan rates for the (a) (8%)rGO/3D ZnO, (b) (16%)rGO/3D ZnO, (c) (32%)rGO/3D ZnO, Calculated C_A values depending on the scan rates (e) in the dark and (e) under UV illumination.



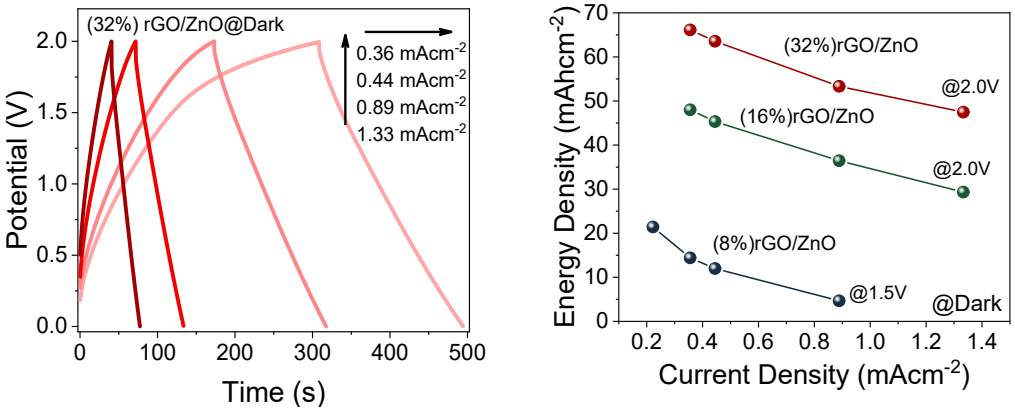


Fig. S5. Dark condition GCD curves at various current densities of the composite PSC devices; (a) (8%)rGO/3D ZnO, (b) (16%)rGO/3D ZnO, (c) (32%)rGO/3D ZnO, (d) comparison of the calculated Ed values depending on applied current densities in the dark condition (The PSC device prepared with the least rGO composition could not reach 2.0V).

Table S2. Values of EC circuit elements of the PSC devices

PSC device	UV condition	R1(Ω)	a1	R2(Ω)	S2 ($\Omega s^{-1/2}$)	C3(F)	R3(Ω)
(8%)RGO/3D ZnO	on	41.09	0.584	62.76	113.8	41.42×10^{-6}	7.76
(16%)RGO/3D ZnO		23.02	0.749	21.74	44.59	0.069	2.1×10^{15}
(32%)RGO/3D ZnO		20.96	0.680	12.41	16.77	0.044	1.3×10^{15}
(8%)RGO/3D ZnO	off	39.20	0.613	128.3	194.2	68.1×10^{-6}	10.01
(16%)RGO/3D ZnO		23.04	0.788	42.18	68.76	0.043	3.1
(32%)RGO/3D ZnO		20.90	0.734	19.34	20.42	0.036	44070

* The variable a ($0 < a < 1$) is the order of the CPE.

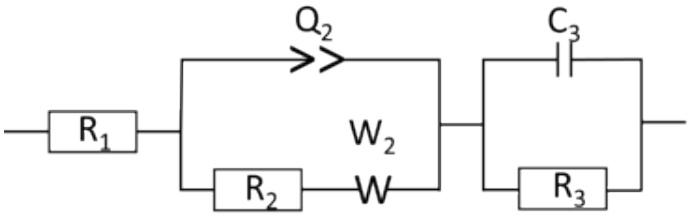


Fig. S6. EC circuit of the PSC devices.

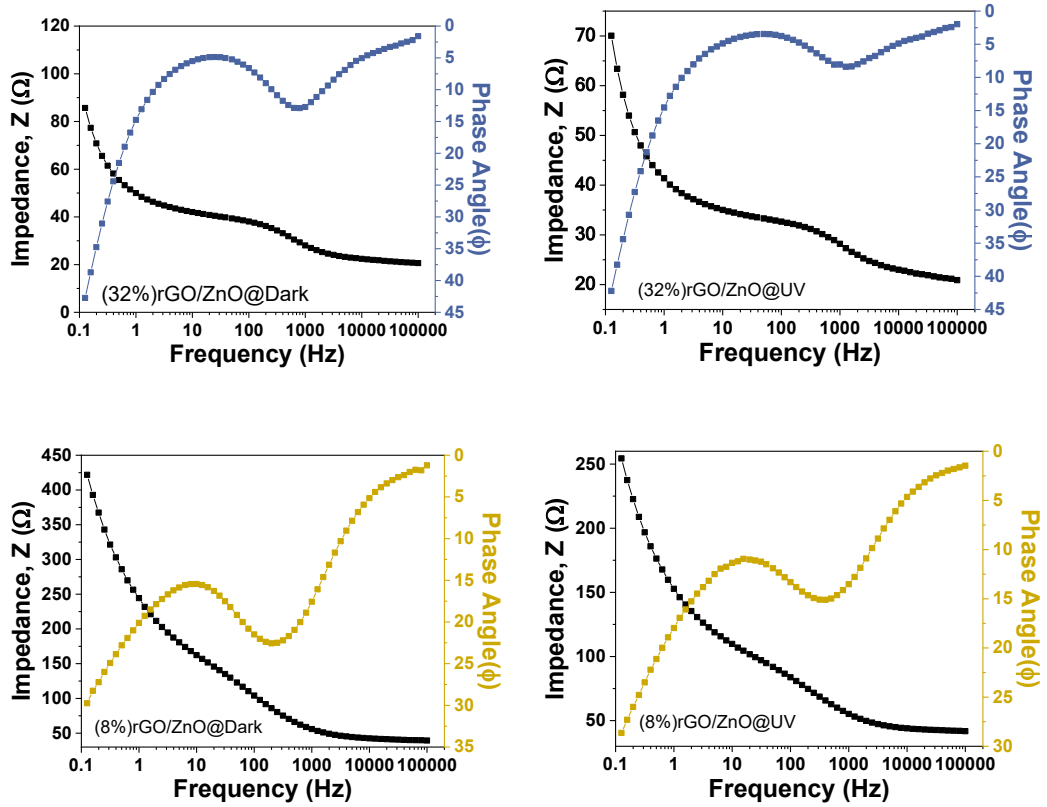
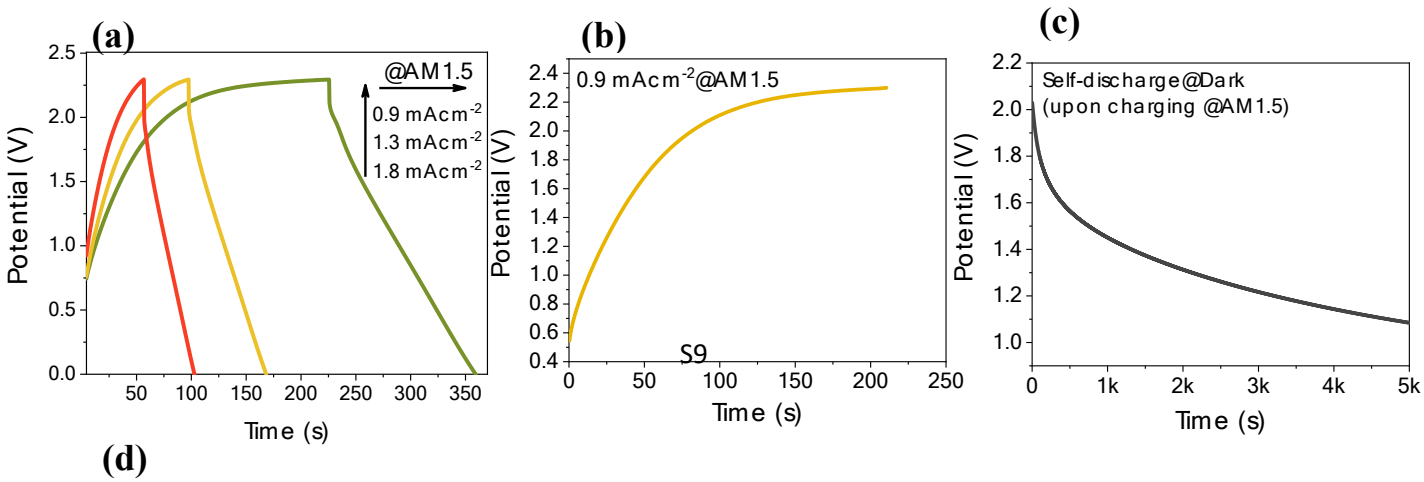


Fig. S7. Bode plots of (8%)rGO/3D ZnO and (32%)rGO/3D ZnO-based PSC devices under UV and dark conditions.



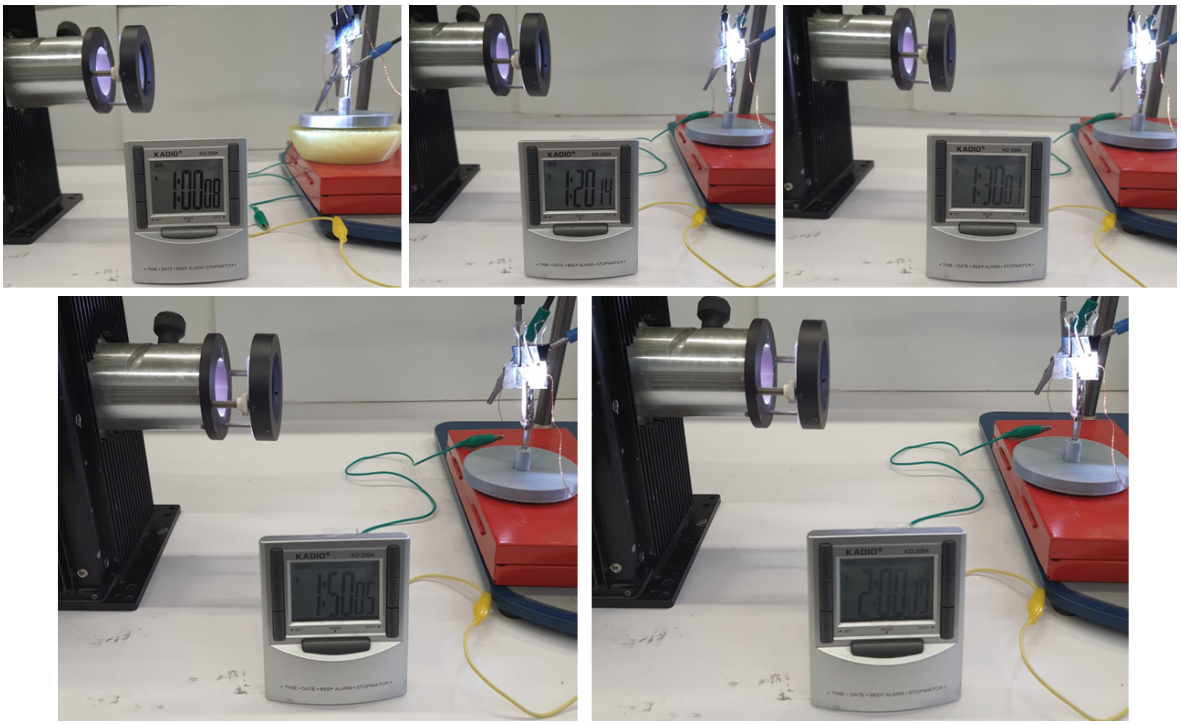


Fig. S8. Proof of concept experiments; (a) GCD curves at various current densities under AM 1.5 light irradiation, (b) Charging at 0.9 $mAcm^{-2}$ under AM 1.5 light irradiation, (c) self-discharge process in the dark after charging at 0.9 $mAcm^{-2}$ under AM 1.5 light irradiation, (c) Photographs of powering a standard digital clock after charging 0.9 $mAcm^{-2}$ (0.2 Ag^{-1}) under AM 1.5 light irradiation.