Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

SUPPORTING INFORMATION

A Three-Dimensional Printed Biomimetic Hierarchical Graphene Architecture for High-Efficiency Solar Steam-Generation

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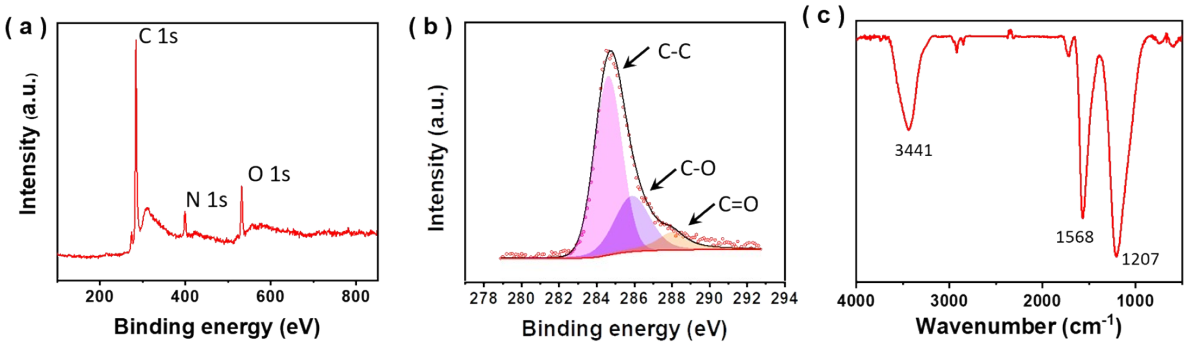

Figure S1. (a, b) C1s and XPS spectra of GOSG. (c) FT-IR spectra of GOSG. (d) XRD spectra of GOSG.

In **Figure S1**a**,** characteristic peaks at 284.5 and 531.7 eV originate from the C1s and O1s on rGO sheets, respectively, and the strong peak at 398.5 eV corresponds to N 1s, confirming the existence of nitrogen-containing functional groups. Some oxygen-containing groups, such as –OH and –COOH peaks, were detected at 286.3 eV and 288.2 eV, respectively (**Figure S1**b). The peaks in the FT-IR spectra located at 1207, 1568, and 3441 cm^{-1} indicate the existence of chemical bonds such as C–N, –NH, and –OH, respectively (**Figure S1**c).

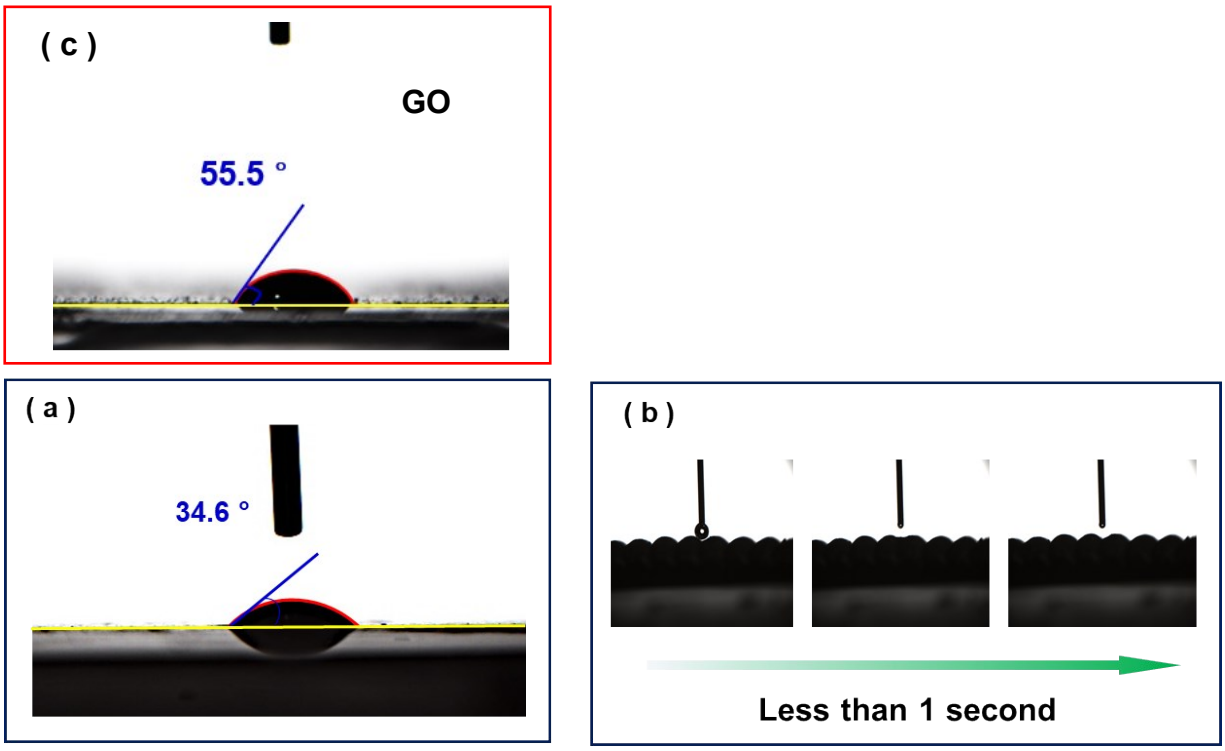

Figure S2. The hydrophilicity of GOSG. (a) Contact angle of the rGO wall in GOSG. (b) Absorption rate test with a water drop on the GOSG surface. (c) Contact angle of the GO wall without chemical modification.

Both modified and unmodified graphene oxide in this work has been characterized via the contact angle, which has been supplemented in the supporting information. While the pure graphene oxide without any modification shows a contact angle of 55.5° and modified rGO shows a contact angle of 34.6° (**Figure S**2a, c), which proved the modified rGO that in GOSG

has better hydrophilicity. A water drop absorbed into the porous surface in much less than 1 second (**Figure S2**b).

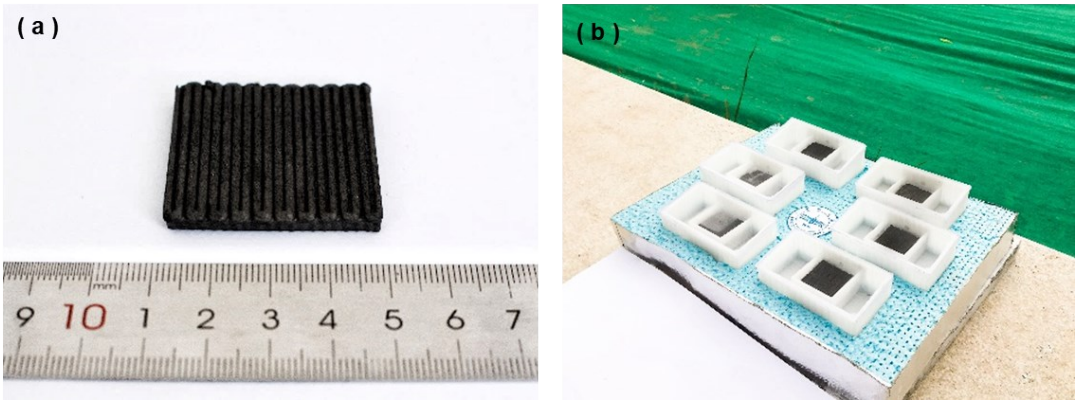

Figure S3. (a) Large-scale GOSG unit for the matrix prototype. (b) Modular prototype assembled into a polystyrene foam as a purification matrix.

The GOSG devices for the large-scale application test were fabricated with an upper structure density of nine ridges, and then assembled in a 3D-printed polylactic acid box. Each prototype was placed in respective positions on a polystyrene foam as a purification unit to easily enable assembly scale-up and convenient maintenance.

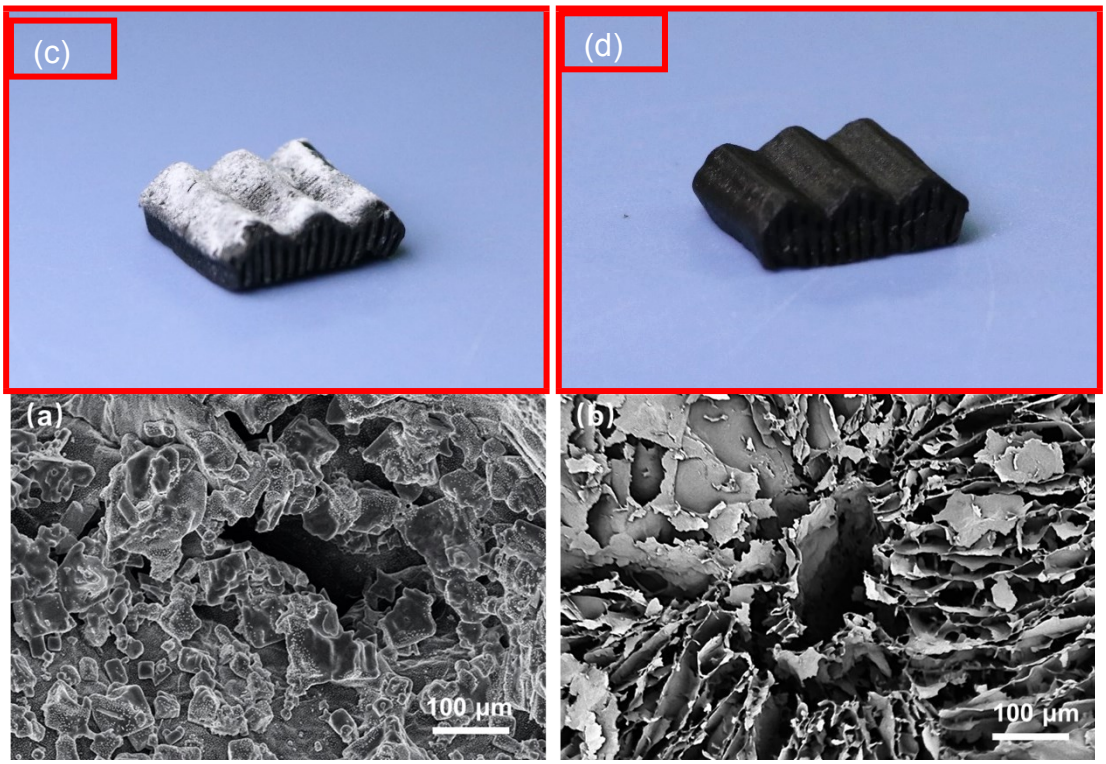

Figure S4. (a) SEM image of the crystallized solution at the bottom channel inlet; the partial blockage of the main channels and sub-pores hindered the water transport. (b) SEM image of the bottom channel inlet treated in-situ with seawater. (c) Optical image of the GOSG salting out because of being illumated 24 h under with a density of 1kW/m^2. (d) Optical image of the GOSG treated in the original seawater by standing still for 2 h.

Some salt separated from solutions to crystallize at the top and bottom, and this hinders the sunlight absorption and water transport due to the partial blocking of the top surface and

main channels and sub-pores. After *in situ* self-cleaning treatments by standing still in the original seawater, the crystallized salts were removed, and a structure with the original high porosity was recovered.

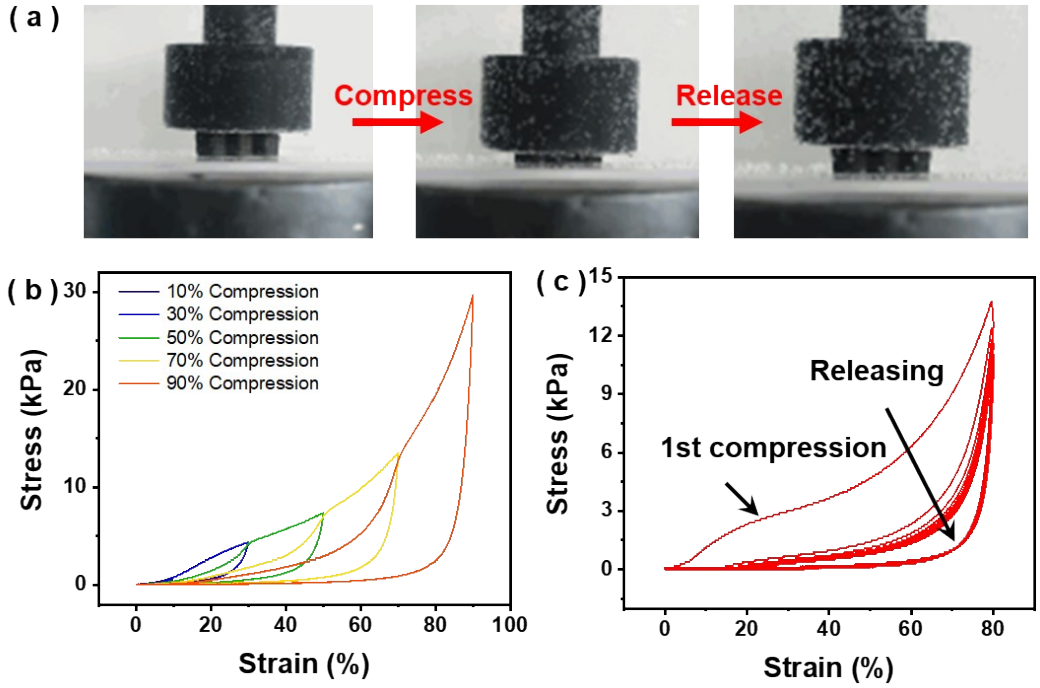

Figure S5. The mechanical properties of GOSG. (a) Digital images showing the recovery of GOSG. (b) Stress–strain curve of GOSG with different compression strains (10%, 30%, 50%, 70%, 90%) during loading–unloading cycles. (c) Loading–unloading curves of GOSG for the first 20 cycles with 80% compression strain.

A highly recoverable compressive strain of 90% and 20 cycles of 80% compressive strain was observed, which indicates that the GOSG can resist possible deformations or fatigue and maintain the structural robustness.

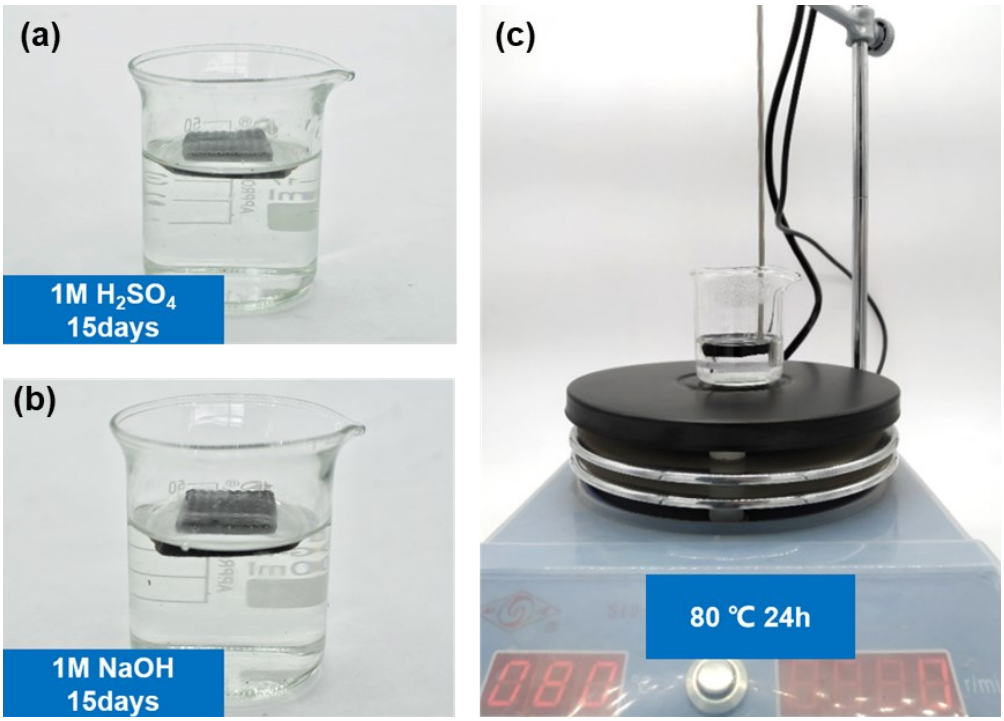

Figure S6. The acid immersion, alkali immersion, and heat durability tests of GOSG. (a) GOSG was immersed in 1M H_2SO_4 solution for 15 days. (b) GOSG was immersed in 1M NaOH for 15 days. (c) GOSG was heated in deionized water under 80°C for 24 h.

The durability was confirmed by the structure retaining its performance after 15 days of the corrosion resistance tests in strong acid (H_2SO_4), after 15 days of alkali (NaOH) solutions (concentration of 1 mol·L^{-1}), and after 24 h heat condition at 80°C in water.

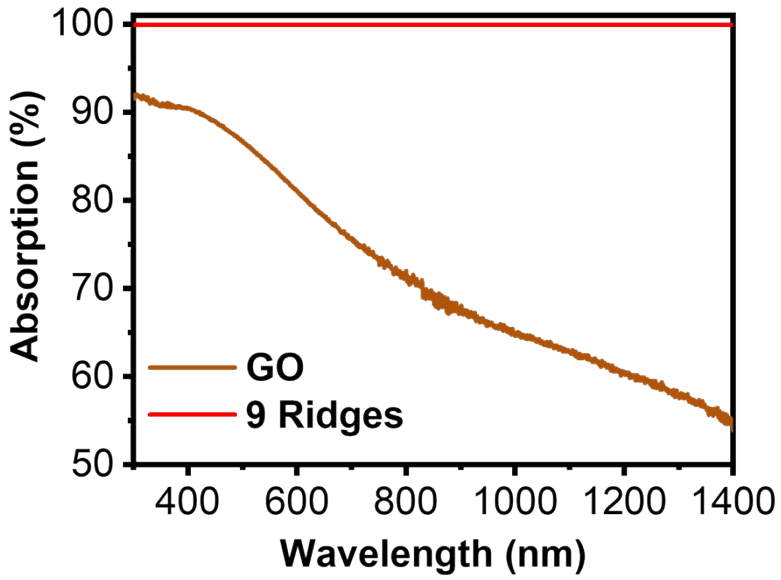

Figure S7. The light absorbing spectrum of 9 bridges GOSG made of modified rGO comparaing to the original GO.

Due to the chemical modification, the light absorbing band of rGO was greatly enlarged than the original GO, which is easily to be explained by the reduction providing more conjugated large π bond.

Geometric transformation parameters of GOSG model

Bridges	Altitude (mm)	Base length (mm)	Hypotenuse length (mm)	Vertex angle (°)	Top area (mm^2)	Amplification factor
0	-	-	-	-	400.0	1.0
3	1.3	6.7	3.6	137.4	429.3	1.1
6	1.3	3.3	2.1	104.1	507.3	1.3
9	1.3	2.2	1.7	81.0	615.6	1.5

Table S1. Geometric transformation parameters of GOSG model with 3, 6, and 9 bridges comparing with the flat sample (0 ridge).

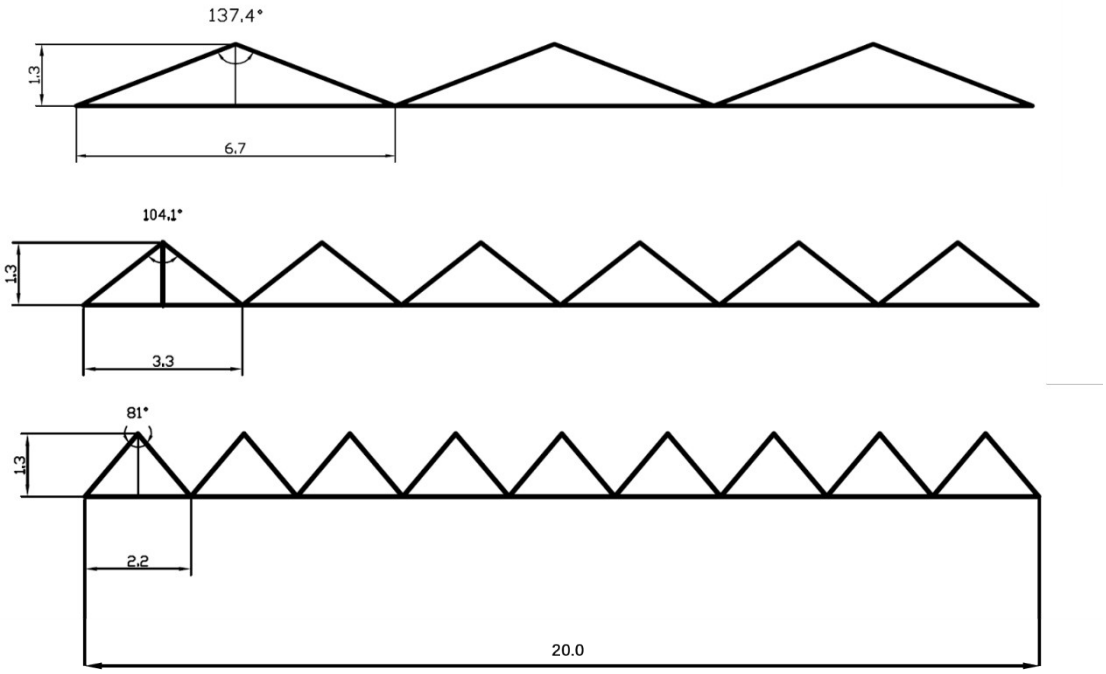

Figure S8. Geometric designing diagram of GOSG with 3, 6, and 9 bridges.

It's important to enlarge the area of the top surface to enhance the sunlight absorption in the same projected area. Theoretically, it's better to have more and sharper ridges to increase the surface area ratio. Therefore different GOSG samples were designed to be vary

at the geometry parameters, so that the top surface area was controlled with a consistent regular which was defined as the *Amplification Factor* (Representing the multiple of surface area increasement than the 0 ridges sample).