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Supporting Information

Robust Graphene Oxide Composite Films via Covalent Cross-linking and

Applicable to Solar Water Evaporators

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Fig S1. AFM image of exfoliated GO nanosheets in GO solution (a) and GO nanosheets in GO/SSA solution (b), and the corresponding specific height of each nanosheets signed by the dashed line in the image.



Fig S2. TGA curves of the GO film, and the prepared GO-SSA-I, GO-SSA-II, GO-SSA-III, GO-SSA-IV composite films, respectively.



Fig S3. (a) XRD results of the GO film, and the prepared GO-SSA-I, GO-SSA-II, GO-

SSA-III, GO-SSA-IV composite films, demonstrating that the interlayer spacing of GO nanosheets gradually expands as the amount of SSA increases. (b) FTIR results of the GO film, and the prepared GO-SSA-I, GO-SSA-II, GO-SSA-III, GO-SSA-IV composite films. It can be clearly observed that the peak position of the -OH within the range of 3600 cm⁻¹ to 3200 cm⁻¹ remains unchanged. It is suggested that there were no hydrogen bonding interactions between GO and SSA in the GO-SSA composite film. In contrast, the characteristic peak at 1038 cm⁻¹ has obviously broadened, demonstrating the formation of a C-O-C covalent bond between SSA and GO nanosheets.



Fig S4. FTIR spectrum of GO film (a) and the GO-SSA composite film (b) respectively. The peak of GO-SSA composite films at ~ 1038 cm⁻¹ is significantly increased, indicating a newly formed C-O-C bonds.

Fig S5. XPS spectra of GO film (a) and the GO-SSA composite film (b) respectively. Compare with the GO film, the S element can be found in the GO-SSA composite film after adding SSA molecules.

Fig S6. The reaction mechanism of GO and SSA through dehydration reaction under heating and acidic conditions. A hydrogen-bonded crosslinked structure is formed between GO nanosheets and SSA in water firstly. And then, most of the water is removed via a vacuum filtration process, the C-O-C covalent bonds are formed after acidic conditions and upon heating process.

Fig S7. Stress-strain curves of (a) GO film, and the prepared (b) GO-SSA-I, (c) GO-SSA-II, (d) GO-SSA-III, (e) GO-SSA-IV composite films, respectively. Each sample represent five parallel results which were shown with lines of different colors.

Fig S8. SEM images characterize the fracture surface of (a) GO film, and the prepared (b) GO-SSA-I, (c) GO-SSA-II, (d) GO-SSA-III, (e) GO-SSA-IV composite films, respectively.

Fig S9. Raman frequency shifts of GO film (a) and the GO-SSA-III composite film (b) during loading/unloading processes. Comparing with the GO film, the GO-SSA-III composite film shows a reversible Raman frequency shift.

Fig S10. The GO-SSA-III composite film (a) and the GO film (b) were ultrasonically treated in the water bath with a frequency of 4.5 kHz under 100 W power. The GO-SSA-III composite film remained intact after ultrasonic treatment for 30 minutes, while the GO film was decomposed and ruptured.

Fig S11. (a) Absorbance spectra of the GO film and the GO-SSA-III composite film performed by UV–vis spectroscopy. (b) Comparison of the XRD result between wet GO-SSA-III composite film and GO film after one water evaporation experiment. The d-spacing of the wet GO film increased to 1.44 nm (calculated from the peak at $2\theta = 6.15$), while the d-spacing of the wet GO-SSA-III composite film was 0.88 nm (calculated from the peak at $2\theta = 10.03$).

Fig S12. The IR thermal images of the GO film and the GO-SSA-III composite film under simulated solar light irradiation $(1.0 \text{ kW} \cdot \text{m}^{-2})$ for 1 hour.

Fig S13. WCA comparison of GO film (a) and the wet GO-SSA-III composite film (b).

Fig S14. Heat flow curves in the process of increasing the temperature from 25 °C to 100 °C of the wet GO film and the wet GO-SSA-III composite film after one water evaporation experiment. The GO-SSA-III composite film was shown a lower initial phase transformation temperature, demonstrating SSA can change the energy requirement for the evaporation of water molecules in the GO-SSA framework.

Fig S15. (a) Evaporation rate of GO film and the GO-SSA-III composite film in pure water cycle 10 times under one sun illumination. It can be clearly observed that the water evaporation rate of GO-SSA composite film remains nearly constant. In contrast, the water evaporation rate of GO film gradually decelerates initially and then experiences a sharp decline starting from the fifth experiment, which is probably attributed to the accumulation of an increasing number of salt deposition within the pores of the GO film. (b) Evaporation rate of GO-SSA-III composite film in 3.5 % and 10 % NaCl solutions cycle 10 times under one sun illumination. The evaporation rate has decreased, which is more noticeable in a 10% NaCl solution.

Fig S16. EDS analysis on the downstream side of the GO-SSA-III composite film (a) and the GO film (b) after a one evaporation experiment with a 3.5 wt% NaCl solution. It is discovered that there was NaCl in the GO film, but not in the GO-SSA-III composite film, which proved that the GO-SSA-III composite film had salt resistance to some extent.

Sample	GO:SSA (wt%)	Tensile strength (MPa)	Toughness (MJ·m ⁻³)	Young's modulus (GPa)
GO	0	128 ± 5.6	2.6 ± 0.2	9.5 ± 0.9
GO-SSA-I	1:5	165 ± 13.9	3.6 ± 0.1	10.2 ± 0.5
GO-SSA-II	1:10	270 ± 37.7	7.2 ± 0.9	11.5 ± 0.4
GO-SSA-III	1:15	418 ± 13.6	11.7 ± 0.5	17.9 ± 2.8
GO-SSA-IV	1:20	162 ± 3.1	4.5 ± 0.3	6.5 ± 1.4

Table S1. Mechanical properties of the GO film, and the prepared GO-SSA compositefilms with different SSA contents.

 Table S2. Comparison of tensile strength and toughness of GO-SSA composite film

 with other GO -based composite films cross-linked through covalent or non-covalent

 interactions.

Sample	Tensile strength (MPa)	Toughness (MJ·m ⁻³)	Reference
GO-PMMA	118	0.1	[18]
GO-PVA	148	2.4	[48]
GO-CNC	490	4	[49]
GO-SL	300	2.2	[50]
GO-Zn ²⁺	142	0.32	[51]
GO-Ca ²⁺	126	0.31	[52]
GO-Mg ²⁺	80.6	0.13	[22]
GO-Al ³⁺	101	0.23	[22]
GO-Cu ²⁺	197	4	[53]
GO-Cd ²⁺	213	2.6	[54]
GO-Fe ³⁺	93	0.6	[55]
GO-BP-AD	389	9.0	[4]
GO-AA-SCMC	305	8.2	[56]
GO-Al ₂ O ₃ -PVA	143	9.2	[57]
GO-MMT-SPVA	250	2.7	[58]
MGO (Before HI reduction)	226	6.2	[59]
GO-SSA-III	418	11.7	Our work

Sample	Water evaporation rate (kg·m ⁻² ·h ⁻¹)	Efficiency (%)	Reference
rGO@fabric	1.54	97.83	[67]
rGO- PAN	1.46	95.4	[68]
SHiCF-GO	1.25	88.9	[69]
PG	1.42	96.6	[32]
NHrG	1.85	95.4	[36]
Co/N-doped GO	2.02		[70]
rGCPP	1.875	84.07	[37]
PPy-V ₂ O ₅ -GO	1.9	92	[71]
GO/ Cu _x S	1.5622	94.61	[72]
PVA/CS/GO-Ag	1.67	93.3	[73]
GO-SSA-III	2.11	93.3	Our work

Table S3. Comparison of water evaporation rates and efficiency of GO-SSA compositefilm with other GO-based evaporators reported in recent years.