Supplementary Information

Tardigrades inspired polyelectrolyte complexation and functional materials

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Supplementary Figure S1. Equipment diagram of probe-type ultrasonic atomization method for preparing PDDA-TFISI/PAA DMSO crystal microparticles.



Supplementary Figure S2. (a) SEM image of the Tar-PEC (PDDA-PAA) microparticles. **(b)** Size analysis of Tar-PEC microparticles by averaging more than 400 particles using SEM image obtained from scanning electron microscopy (a). **(c)** Size distribution of Tar-PEC particles by light scattering measurement (Mastersizer 2000) of PEC particles dispersed in water, (d) XRD patterns of PAA, PDDA-TFSI and PDDA-PAA Tar-PEC particles. Please note: XRD was conducted by Bruker D8 ADVANCE equipment (angular range of 5°-80°, scanning speed: 10°/min).



Supplementary Figure S3. Effect of soaking time in EtOH/NH₃ solvent (pH ~11.2, T_0 =-30 °C) on the sulfur and fluorine elements content of PDDA-PAA complex particles. Note: Since counter-ions were quantitatively released as a result of complexation, the electrostatic complexation degree (ECD) of PDDA-PAA Tar-PEC could be calculated by the ratio of TFSI release [Ref: Nat. Commun. 2014, 5, 5293.] during the freeze-thaw complexation. ECD was calculated by fluorine element changes during the complexation.

$$ECD\% = \frac{A_F - A_{Ft}}{A_F}\%$$

Above, A_F is the atomic ratio of fluorine element in PDDATFSI-PAA blend before immersing in EtOH/NH₃ bath, A_{Ft} is the atomic ratio of fluorine at varied immersion time. The term $[A_F-A_{Ft}]$ represents the amount of TFSI anions that have released, corresponding to the amount of PDDA monomers that have complexation with negatively charged PAA. A_F and A_{Ft} were determined by XPS analysis (F element).



Supplementary Figure S4. Dynamic thawing actuated complexation processes of PDDA-TFSI/PAA DMSO crystals in EtOH/NH₃ solutions (pH ~11.2) with different



initial temperatures (from top-down direction: -30 °C, 2 °C, 50 °C). Scale bar, 0.2 mm.

Supplementary Figure S5. A proposed mechanism of the temperature-dependent architecture adjustment of PDDA-PAA Tar-PEC particles during the freeze-thaw process.



Supplementary Figure S6. (a) Particle size distribution of CaCO₃ microparticles. **(b-c)** SEM images of the as prepared Tar-PEC (PDDA-PAA) microcapsules. The preparation of CaCO₃ microparticles: in a typical experiment, 0.33 M Na₂CO₃ solution was rapidly poured into an equal volume of 0.33 M solution of CaCl₂ at room temperature, and after intense agitation (1000 rpm) on a magnetic stirrer, the precipitate was filtered off, thoroughly washed with pure water, and dried in air.



Supplementary Figure S7. (a) Digital camera pictures of as prepared Tar-PEC (PDDA-PAA) membrane. **(b)** Structure modulation of Tar-PEC (PDDA-PAA) membranes by tuning the starting PDDA-TFSI/PAA concentrations before the freeze-thaw complexation.



Supplementary Figure S8. (a) Surface temperature changes of different samples under one sunlight irradiation (1 kW m⁻²). PEC-M: PDDA-PAA membrane, PEC-CM: PDDA-PAA-CNT membrane. C-PVDF: CNTs coated PVDF membrane. (b) Infrared photos of different samples under one sunlight irradiation (1 kW m⁻²) at 200 s.

Materials	$\Delta m (kg m^2 h^{-1})$	Efficiency	Refs
Tar-PEC hybrid membranes	1.55	82 %	This work
RGO-SA-CNT	1.62	83 %	[1]
Graphene foam	1.6	91.4 %	[2]
Graphene oxide based membrane	1.45	80 %	[3]
PPy nanosheets	1.38	92 %	[4]
biochar-based absorbers	1.21	80 %	[5]
SWNT-MoS2 film	1.2	81 %	[6]
Polymer foam	1.17	80.5 %	[7]
PIL-derived carbon membranes	1	75 %	[8]
Plasmonic woods	1	68 %	[9]
Au/D-NPT	1	64 %	[10]
Wood/CNTs	0.95	65 %	[11]

Supplementary Table S1. Data collection for Figure 8d in the main text.

polyurethane sponges	0.83	54 %	[12]
Black gold membranes	0.67	42 %	[13]
Honeycomb graphene foam	0.5	40 %	[14]

Supplementary references of Table S1

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