# Setaria viridis-inspired hydrogels with multilevel structures for all-day fresh water harvesting <br> Xin Su, ${ }^{\text {a,b }}$ Dezhao Hao, ${ }^{c}$ Pei Li, ${ }^{\text {a }}$ Ming Yang, ${ }^{a}$ Xinglin Guo, ${ }^{* a}$ Xicheng Ai, ${ }^{* b}$ Tong Zhao, ${ }^{\text {a }}$ and Lei Jiang ${ }^{\text {c }}$ 

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## Supplementary methods

Preparation of PVA/GO hydrogel: To prepare the gel precursor solution, PVA (1 g), and graphene oxide solution ( $9 \mathrm{~mL}, 3 \mathrm{wt} \%$, dispersed using high speed shears) were dissolved in hot water for 2 hours, $\mathrm{HCl}(50 \mu \mathrm{~L}, 1.2 \mathrm{M})$ and glutaraldehyde ( $30 \mu \mathrm{~L}, 50$ $\mathrm{wt} \%$ solutions were mixed together. The gel was then frozen-thawed and the cycle is repeated 3 times. After being freeze-dried for 48 h and then treated in an oven at 60 ${ }^{\circ} \mathrm{C}$ for 2 hours to prepare the hydrogel.

Preparation of PVA-PEG/GO hydrogel: PVA (1 g), PEG600(0.5 g), and graphene oxide solution ( $9 \mathrm{~mL}, 3 \mathrm{wt} \%$, dispersed using high speed shears) were dissolved in hot water for 2 hours. $\mathrm{HCl}(50 \mu \mathrm{~L}, 1.2 \mathrm{M})$ and glutaraldehyde ( $30 \mu \mathrm{~L}, 50 \mathrm{wt} \%$ solutions were mixed together. The mixture of gel precursor solution was transferred into a PTFE mold. Then the mold was placed on the surface of liquid nitrogen for 10 min for directional freeze casting from the bottom to the top. Finally, the hydrogel was obtained by freeze-drying and then treated in an oven at $60^{\circ} \mathrm{C}$ for 2 hours.

Preparation of PVA-PEG-CS/GO hydrogel: The concentration of the chitosan solution (Macklin) is $30 \mathrm{mg} \mathrm{mL}^{-1}$. Typically, 2 mL of glacial acetic acid was added into 97 mL of de-ionized water, and then 3 g of chitosan was dispersed in the suspension under vigorous stirring. In a typical example: $\operatorname{PVA}(1 \mathrm{~g}), \operatorname{PEG}(0.5 \mathrm{~g})$, chitosan solution (PVA-PEG-CS1/GO, 1 g ) (PVA-PEG-CS2/GO, 1.5 g ), and graphene oxide ( $9 \mathrm{~mL}, 3 \mathrm{wt} \%$ ) were dissolved in hot water for 2 hours, $\mathrm{HCl}(30 \mu \mathrm{~L}$, 1.2 M) and glutaraldehyde ( $80 \mu \mathrm{~L}, 50 \mathrm{wt} \%$ solutions were mixed together. The mixture of gel precursor solution was transferred into a PTFE mold. Then the mold was placed on the surface of liquid nitrogen for 30 min for directional freeze casting from the bottom to the top. Finally, the hydrogel was obtained by freeze-drying and then treated in an oven at $60^{\circ} \mathrm{C}$ for 2 hours to prepare the hydrogel.


Figure S1. SEM image of Setaria viridis, the tip with an apex angle and dense oriented barbs, the middle with gradient grooves.


Figure S2. SEM image of hydrogels. (a) PVA/GO hydrogels after 3 freeze-thaw. (b) PVA-PEGCS/GO hydrogels after 3 freeze-thaw. (c,d) PVA-PEG-CS/GO hydrogels after ice template method.



Figure S3. Two-dimensional SAXS plots of BSVH and BSV-VAH hydrogels.


Figure S4. Mechanical properties of hydrogels and Raman analysis. (a) Compression properties of different hydrogels. It was worth noting that the PVA/GO solution cannot be made into a hydrogel after a single directional freezing. (b) Photos of hydrogel deformation during compression and photos of gelation of different hydrogel samples during freezing-thawing. (c) Dynamic mechanical analysis of the storage modulus $\left(\mathrm{G}^{\prime}\right)$. The results demonstrate that PEG and CS penetrates the molecular meshes among the PVA/GO network. (d) The Raman curves of hydrogels.


Figure S5. Water content, DSC analysis, and dark evaporation rate of different samples. (a,b) Dark evaporation rate and equivalent enthalpy of different hydrogels. (c) The water content after equilibration in pure water. (d) DSC curves showing the melting behavior of water in different frozen hydrogels.

