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## **Supporting Information**

Hydrovoltaic Energy Harvesting at Ionic Polymer-Hydrogel-Carbon

Composites via Moisture Flow

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Fig. S1 (A) Materials used to fabricate IPHC consisting of a glass slice, carbon paper, Nafion polymer membrane, pNIPAm hydrogel nanoparticles, pored carbon paper with artificial holes, and Ni/Ag alloy metal tapes. (B) Digital image of an IPHC device.



Fig. S2 Electric output of an IPHC device connecting to different load resistances. In this experiment, the device was continuously exposed to the moisture flow while different load resistances were connected in turn from large to small, which is indicated by X-axis of time. Open circuit (OC) condition is shown for the period between two resistances were changed.



Fig. S3  $V_{oc}$  of a special IPHC sample in response to moisture flow. Blue region represents the supply of moisture flow from 30s to 120s, and light blue region indicates the reduction of voltage response after fully swollen of the pNIPAm hydrogel. Specifically, this sample was prepared by carrying out the polymerization of pNIPAm on the pored CP directly, which features an extremely thin pNIPAm layer. Insert shows digital image of the pored CP coated with pNIPAm.



Fig. S4  $V_{oc}$  of an IPHC under continuous moisture flow until its saturation. Blue region shows the supply of moisture flow and light blue region indicates the saturation of the device even the moisture flow lasts after 600s. (The measurement uncertainty in terms of  $V_{oc}$  of *ca*. ±20% could be observed when the IPHC sample was initially exposed to the moisture flow.)



Fig. S5 Saturation time of IPHCs with different layer thickness of pNIPAm.



Fig. S6 Charge density difference plots for H adsorbed on (A) graphene, (B, C, and D) pNIPAm-graft-graphene at different absorption sites on the condition without water molecule. The yellow and cyan distribution corresponds to charge accumulation and depletion, respectively. The isosurface value equals to 0.002e  $Å^{-1}$ .



Fig. S7 Additional adsorption configurations of a H on (A) graphene with H<sub>2</sub>O molecule, (B) pNIPAm-graft-graphene with H<sub>2</sub>O, and (C) pNIPAm-graft-graphene, and corresponding adsorption energies.



Fig. S8  $E_a$  of graphene and isoheptane-graft-graphene to H on the condition with (blue) and without (green) H<sub>2</sub>O.  $E_a$  of meta-stable adsorption configurations are presented as dashed lines. Inset shows the structural formula of isoheptane molecule.



Fig. S9 Adsorption configurations of a H on (A) isoheptane-graft-graphene and (B) isoheptance-graft-graphene with  $H_2O$ , and corresponding adsorption energies.



Fig. S10 Surface potential of CP and pNIPAm-graft-CP which were then subjected to moisturizing, baking, and drying via nitrogen gas in proper order.



Fig. S11 Digital images of a sample with one-half pNIPAm-graft-CP (left) and one-half pristine CP (right) at (A) dry state and (B) wet state.



Fig. S12 *V*<sub>oc</sub> of an IPHC in response to the change of RH for the entire environmental chamber and corresponding RH.



Fig. S13  $V_{oc}$  of an IPHC in response to the supply of dry air and corresponding flow rate.



Fig. S14  $V_{oc}$  of an IPHC that was exposed to dry air at a flow rate of 3.0m s<sup>-1</sup> from 11s to 90s, and then moisture flow from 39s to 110s. The schematic diagram illustrates the experimental setup where the moisture was driven into the IPHC device by forced convection during 39-90s.



Fig. S15 (A)  $V_{oc}$  of two IPHC devices connected in series. (B)  $J_{sc}$  of two IPHC devices connected in parallel.



Fig. S16 (A)  $V_{oc}$  of an IPHC device comprising poly(N-isopropylacrylamide) (pNIPAm) and 2-aminoethylmethacrylate hydrochloride (AEMA) co-polymerized hydrogel microparticles (pNIPAm-AEMA) as the moisture absorbent layer. Inset shows the size distribution of asprepared pNIPAm-AEMA. (B)  $V_{oc}$  of an IPHC device comprising polypyrrole (PPy) hydrogel nanoparticles as the replacement of pNIPAm. Inset shows the digital image of PPy nanoparticles.



Fig. S17 Particle size distribution of pNIPAm hydrogel nanoparticles.



Fig. 18 Equivalent circuit of the IPHC device.



Fig. S19 (A)  $V_{oc}$  of an IPHC device in response to moisture flow at specific cycles. (In each cycle, the device was moisturized to its saturation and then baked to dry.) (B) Stability in terms of built-in voltage and voltage output for 12 cycles.