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Supporting Information

Stable Hydrophobic Ionic Liquid Gel Electrolyte for Stretchable Fiber-Shaped Dye-Sensitized Solar Cell

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Experimental section

Preparation of stretchable CNT fiber. A CNT sheet was first drawn from a spinnable CNT array and then wrapped onto a pre-stretched commercial polyurethane elastomer fiber (diameter of 600 μm). The experimental setup included a motor to rotate the fiber substrate and a translation stage to move the CNT array.[S1] The rotating speed was 60 rounds per minute and the moving speed of the stage was 10 mm/min. For a CNT sheet with a width of 5 mm, thirty layers of CNT sheets were wound on the elastomer fiber.

Synthesis of electrolytes. Liquid electrolyte, DHS-Et23, was ordered from HEPTACHROMA and was mainly composed of acetonitrile, ionic liquid, 4-tert-butylpyridine, LiI and I_2. MPN gel electrolyte was prepared according to the previous report,[S2] and it contained 0.1 M LiI, 0.05 M I2 , 0.5 M 4-tert-butylpyridine, 0.5 M ionic liquid 1-propyl-2,3-dimethylimidazolium iodide and 10 wt% Poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP) with MPN as the solvent. The IL gel electrolyte was prepared by adding the same amount of 1-butyl-3-methylimidazolium bis(trifluoromethanesulfonyl)imide (BMITFSI) to the 10 wt% MPN solution of PVDF, followed by heating to 100 °C for 24 h in an oven with flowing air to fully remove the MPN and forming a BMITFSI solution of PVDF. The other components were added as the same quantity to the MPN gel.

Fabrication of stretchable DSSCs. The fabrication details were reported elsewhere.[S1] Briefly, a titanium wire was firstly wound onto a steel rod with a diameter of 600 μm to form a spring structure (Figure 2a). The titanium wire was anodized to grow titania nanotube array on the surface at 60 V, followed by annealing in air at 500 ℃ for 1 h. The modified titanium wire was then treated by a titanium tetrachloride solution under 70 ℃ for 30 min, annealed again at 450 ℃ for 30 min and immersed into 0.3 mM N719 dye solution for 12 h and after cooling to 120 ℃. The modified spring was inserted with a CNT fiber and put into a heat-shrink tube, followed by injection of the electrolyte. For the MPN and IL gel electrolyte, they were heated to form liquids prior to the injection. Gels were formed after cooling down to room temperature. To obtain encapsulated DSSCs, the heat-shrink tube was treated at 100 ℃ with both ends sealed by a UV-cured epoxy resin. For the DSSC without encapsulation, the heat-shrink tube would be removed after the gel was formed (Figure S3).

Characterizations. The structures were characterized by scanning electron microscopy

(Hitachi FE-SEM S-4800). The current-voltage curves were recorded under 100 mW cm^{-2} AM1.5 simulator (Oriel-Sol 3A 94023A equipped with a 450 W Xe lamp and an AM1.5 filter). EIS measurements were performed on a CHI-660 electrochemical workstation.

References:

[S1] Z. Yang, J. Deng, X. Sun, H. Li, H. Peng, *Adv. Mater.* **2014**, *26*, 2643-2647.

[S2] S. Pan, H. Lin, J. Deng, P. Chen, X. Chen, Z. Yang, H. Peng, *Adv. Energy Mater.* **2015**, *5*, DOI 10.1002/Aenm.201401438.

Supporting videos

Video S1. Ionic liquid (IL) gel electrolyte being bent to demonstrate a high flexibility.

Video S2. A fiber-shaped dye-sensitized solar cell being stretched to 25% and then released.

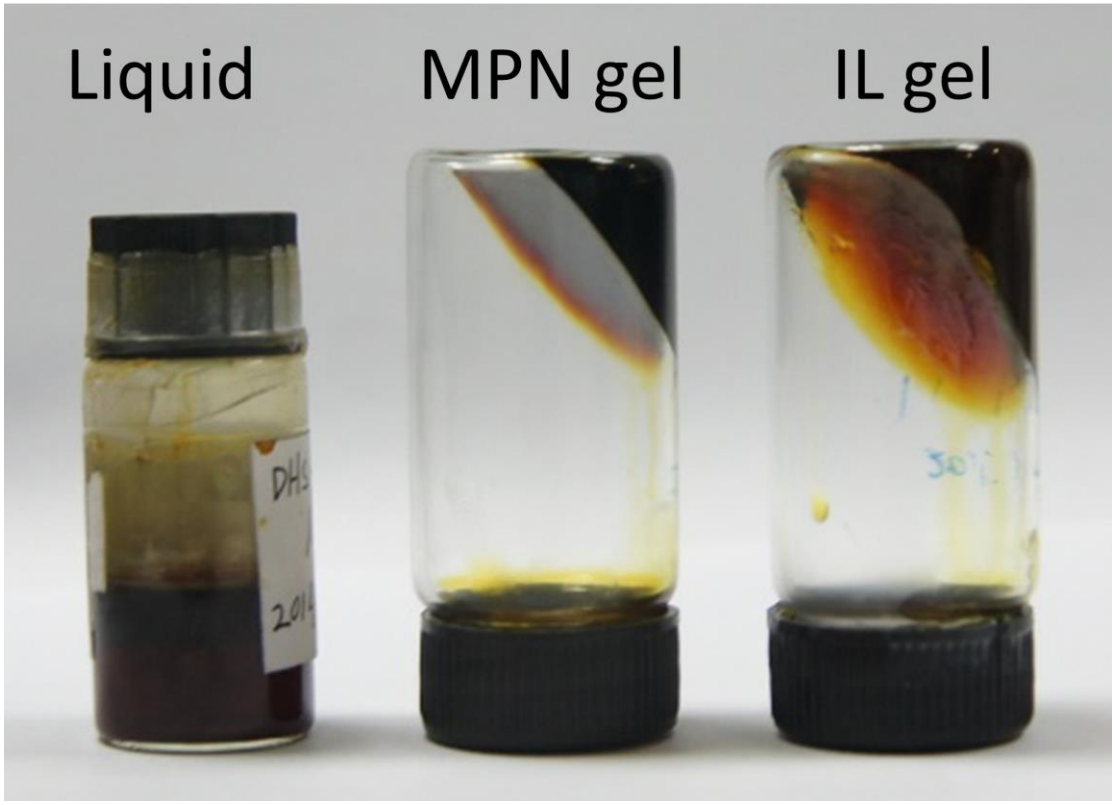

Figure S1. Photographs of three gel electrolytes.

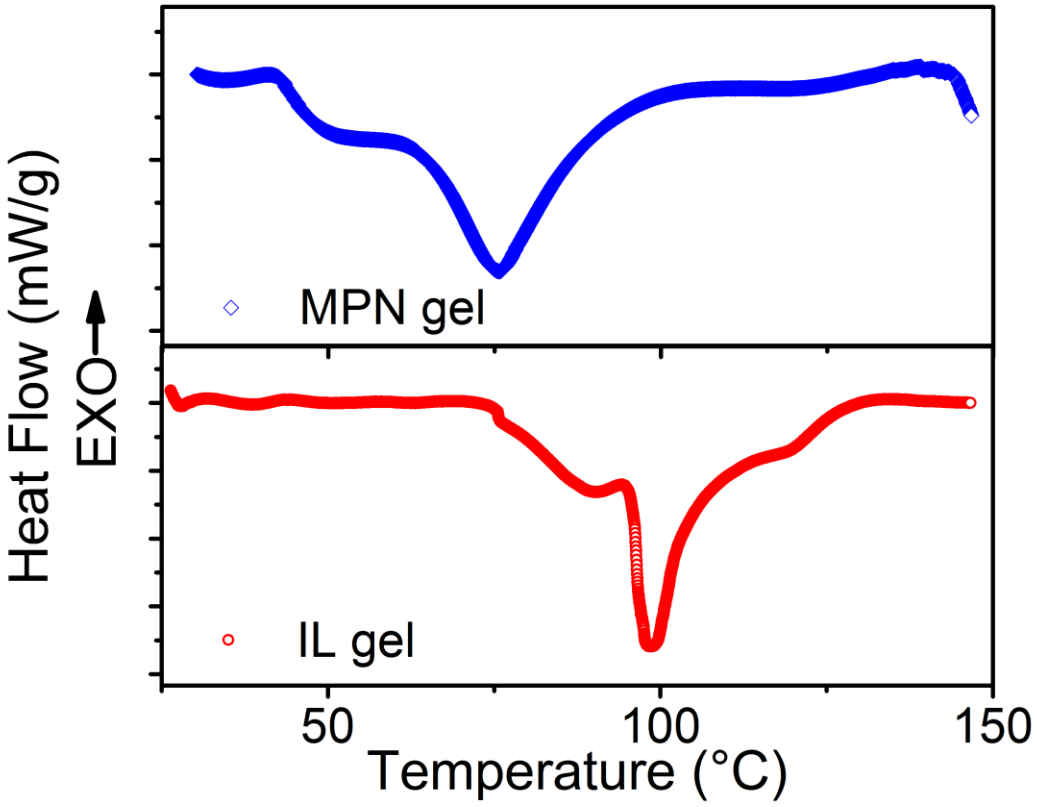

Figure S2. Differential scanning calorimeter (DSC) curves of MPN and IL gels with gelling points of 75 and 98 ℃, respectively.

Figure S3. Dye-sensitized solar cells (DSSCs) fabricated from the IL gel electrolyte with (**a**) and without (**b**) encapsulation.

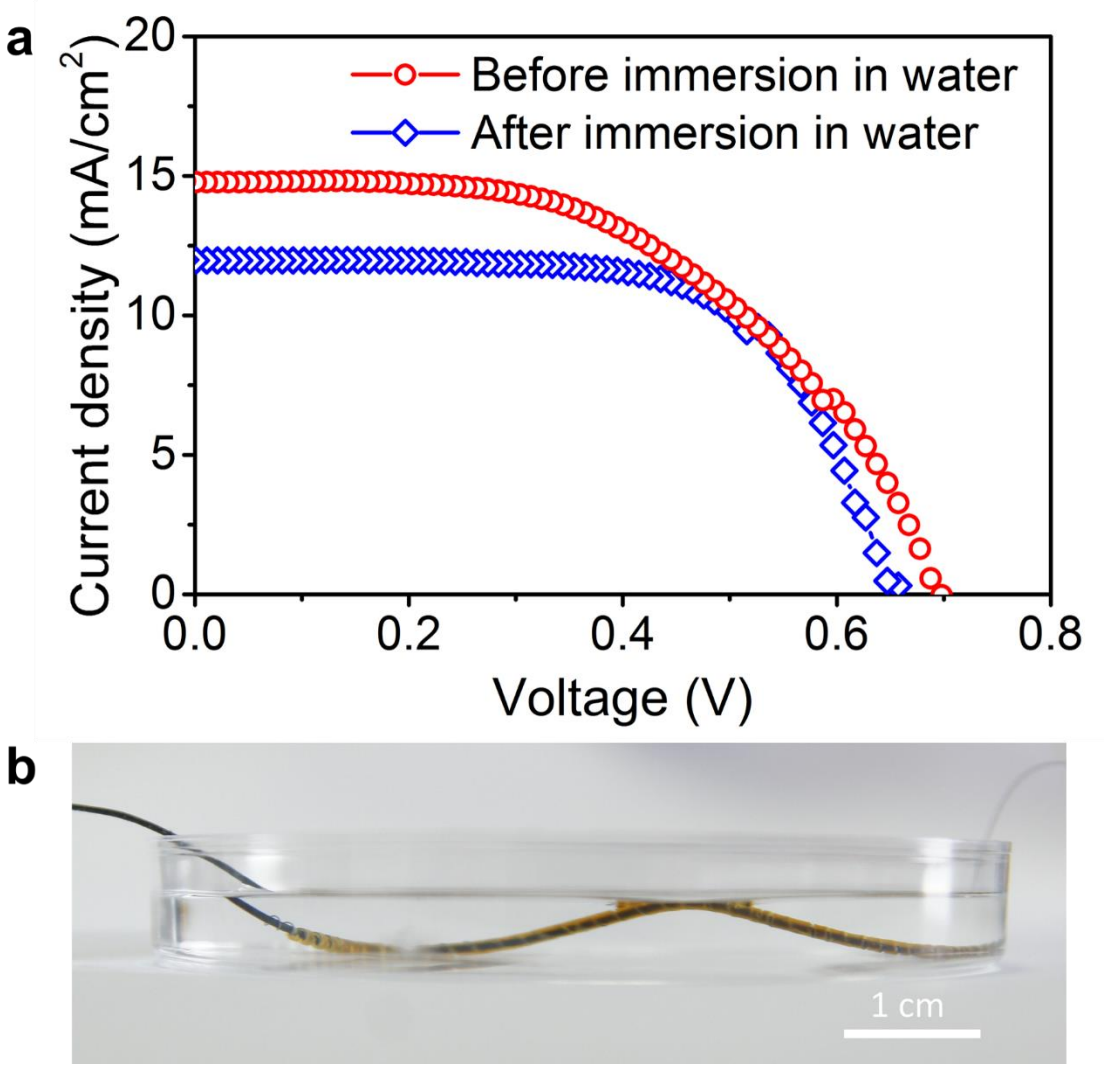

Figure S4. a. *J-V* curves of the DSSC derived from the IL gel electrolyte before and after immersion in water. **b.** Photograph of a DSSC immersed in water at 25 ℃ for 1 min.

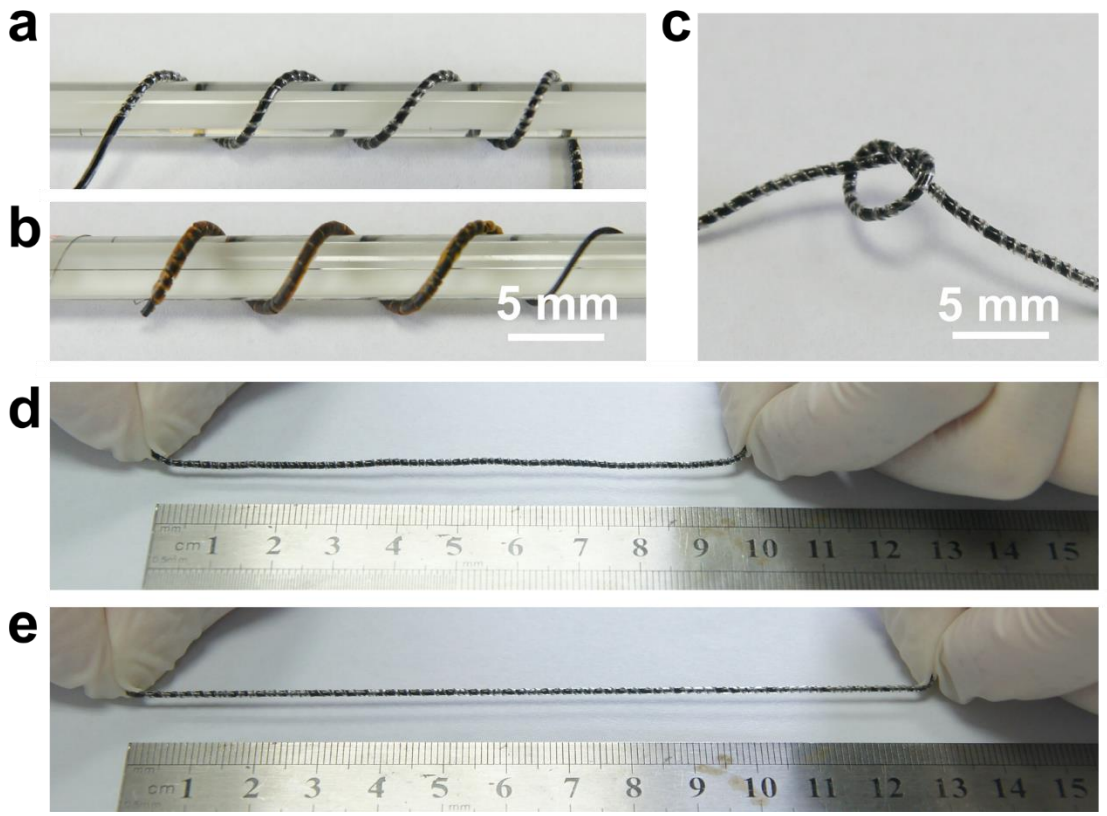

Figure S5. Photographs of a fiber-shaped DSSC fabricated from the IL electrolyte being wound on glass rods with (**a**) and without (**b**) encapsulation. **c.** Being knotted. **d.** At a relaxed state. **e.** Being stretched

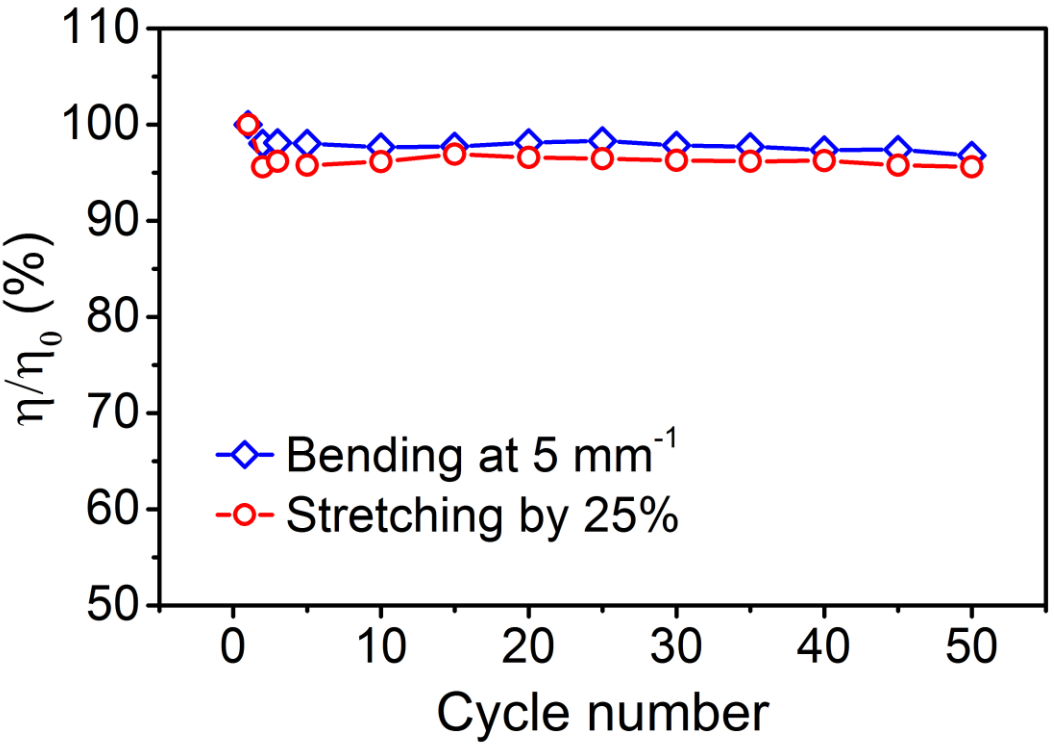

Figure S6. The change of power conversion efficiency in a DSSC with IL gel electrolyte after bending and stretching for 50 cycles.