Transparent and adhesive carboxymethyl cellulose/polypyrrole hydrogel electrode for flexible supercapacitor

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KPS induced aggregation state change of CMC/PPy fibers

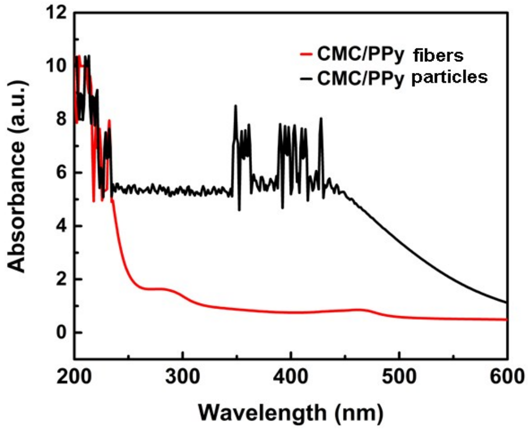

Fig. S1 The UV-Vis absorption spectra of CMC/PPy fibers and uniformly distributed CMC/PPy particles after processed with KPS.

In order to investigate the unique visible light transmission and ultraviolet (UV) shielding optical characteristics to PPy hydrogel electrode, the CMC/PPy fibers and the CMC/PPy particles were analyzed. The KPS (5 mg) was directly added in the CMC/PPy fibers (PPy of 1.0 vol%) aqueous solution to prepare the uniformly distributed CMC/PPy particles. As illustrated in **Fig. S1**, the uniformly distributed CMC/PPy particles with small size possessed the excellent absorption performance in UV region.

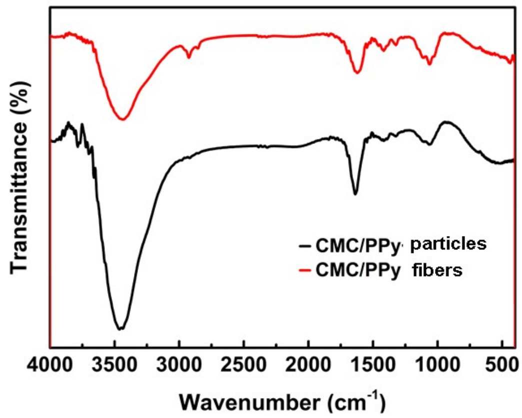

Fig. S2 The FITR spectra of CMC/PPy fibers and uniformly distributed CMC/PPy particles after processed with KPS.

Before the FTIR and analysis, the CMC/PPy fibers and CMC/PPy particles solution after incubation with KPS were freezing-dried. As illustrated in **Fig. S2**, compared with CMC/PPy fibers, the chemical structure of CMC/PPy particles after processed with KPS was not difference. During the incubation process, only the aggregate state of CMC/PPy fibers changed, and the structure did not change.

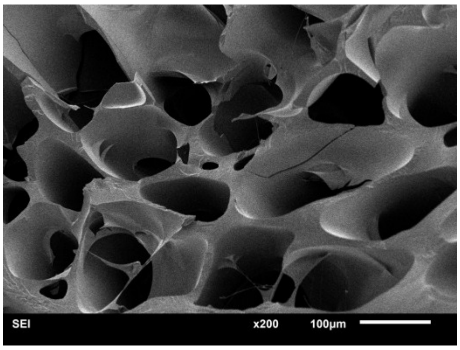

Fig. S3 The SEM photography of dehydrated PAAm/CMC/PPy hydrogel.

After in-stiu formation of CMC/PPy particles, the dehydrated PAAm/CMC/PPy hydrogel was observed by the SEM. In **Fig. S3**, the CMC/PPy particles owned a good affinity for the network inside the hydrogel, which combined with polymer network to fabricate the conductive pathway and improve conductivity. And the interconnected porous structure of PAAm/CMC/PPy hydrogel was observed.

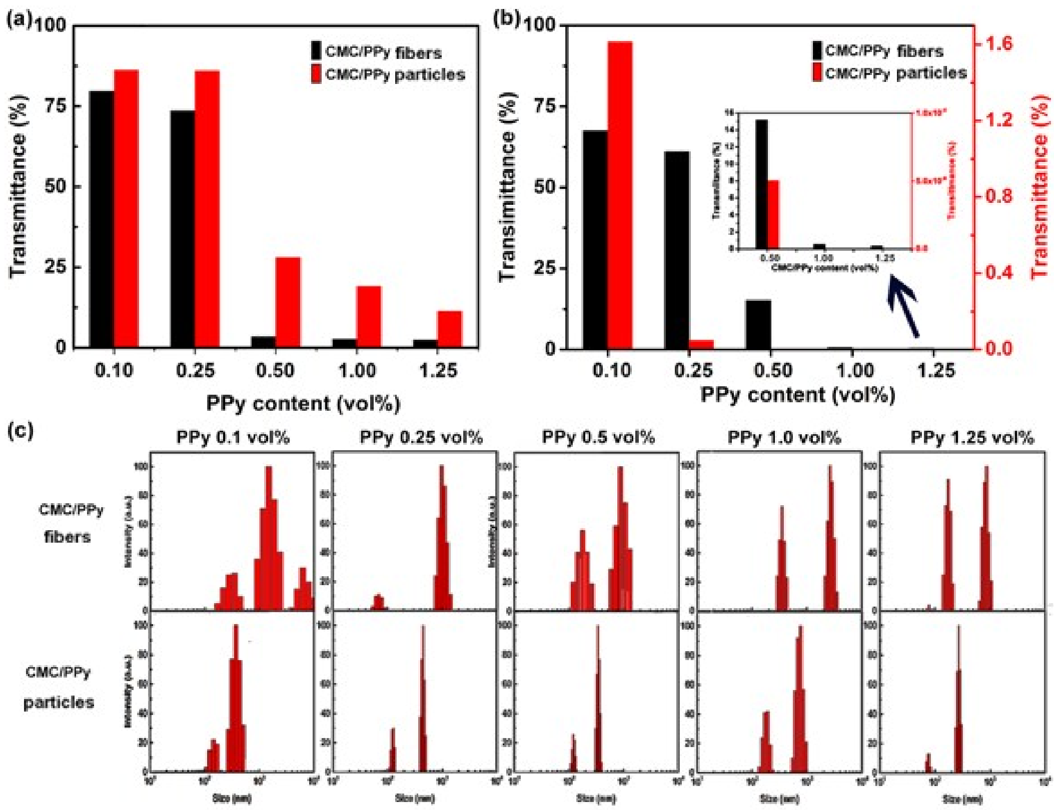

Fig. S4 Transmittances of CMC/PPy fibers and CMC/PPy particles after processed with KPS at (a) 660 nm and (b) 365 nm; (c) DLS of CMC/PPy fibers and CMC/PPy particles with the various PPy contents

The unique visible light transmission and ultraviolet (UV) shielding optical characteristics of PPy hydrogel were deprived from the in-situ formed uniformly distributed CMC/PPy inside the hydrogel. The optical performance of CMC/PPy fibers and CMC/PPy were analyzed by UV-Vis absorption spectra. As shown in **Fig. S4a and b**, the UV-shielding behavior of uniformly distributed CMC/PPy particles was excellent obvious. Moreover, the sizes of CMC/PPy fibers and CMC/PPy particles with various PPy contents were quantitatively analyzed by DLS. In **Fig. S4c**, the particle sizes of CMC/PPy particles changed smaller and size was lower than 660 nm. Therefore, the PAAm/CMC/PPy hydrogel possessed the visible transparency.

The dynamic mechanical property of PAAm/CMC/PPy hydrogel

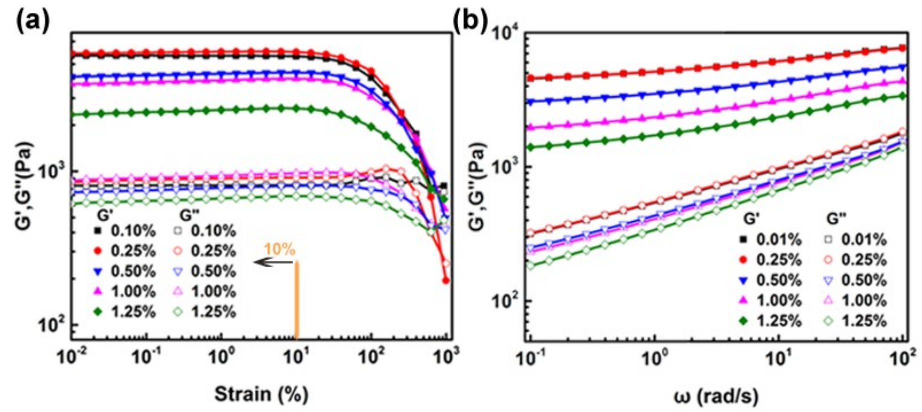

Fig. S5 The a) amplitude sweep and b) frequency sweep curves of PAAm/CMC/PPy hydrogel with various PPy contents.

The dynamic mechanical properties of hydrogel were researched by rheological measurements. In **Fig. S5a**, there existed a negligible weak strain overshoot phenomenon for PAAm/CMC/PPy hydrogel which derived from H-bonds. According to strain sweep curves, the linear region was determined as the stain of 10%, which did not depend on strain. Then, the dynamic gel network relaxation property was analyzed by frequency sweep measurements in **Fig. S5b**. The storage modulus (G') and loss modulus (G") were decreased as the frequency declined indicating the progress of relaxation. Moreover, the linear relationship between G" and frequency reflected the typical feature of covalent crosslinking network. Interestingly, the G' of PAAm/CMC/PPy hydrogel containing PPy of 0.25% showed the maximum value consistent with tensile and compression conclusions. The G' of hydrogel depended on the content of PPy in that the addition of PPy could inhibit the monomer polymerization resulting in the improved viscosity.

The conductivity of PAAm/CMC/PPy hydrogel

The conductivity of PAAm/CMC/PPy hydrogel sample (25 mm×4 mm) was measure by a two electrode system (Solartron 1260/1287 electrochemical workstation). The conductivity of hydrogel sample was calculated as following:

$\sigma = L/RS$

where L, S and R was the length of hydrogel sample, the conductive area and resistance of hydrogel, respectively.

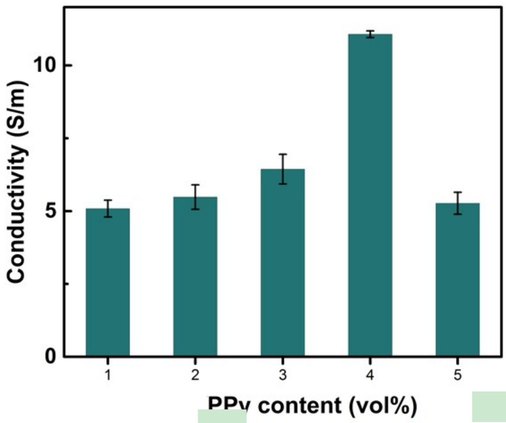

Fig. S6 The electrical conductivity of hydrogel with various PPy contents.

The electronic conductivity of PAAm/CMC/PPy hydrogel with various PPy contents was analyzed. In **Fig. S6**, as the PPy content increased, the conductivity increased followly declined, and the maximum value attained 11.07 S/m.

Table S1. The recipes of the PAAm/CMC/PPy hydrogels.

Hydrogel	PPy (vol%)	CMC (wt%)	AAm (g)	MBA (wt% of AAm)	KPS (wt% of AAm)	TMEDA (uL)
PAAm/CMC/PPy	0.10	0.75	4	1.5	7.5	10
	0.25	0.75	4	1.5	7.5	10
	0.50	0.75	4	1.5	7.5	10
	1.00	0.75	4	1.5	7.5	10
	1.25	0.75	4	1.5	7.5	10

Table S2. The conductivities, stresses and specific capacitances for recently reported conductive hydrogel.

Name	Conductivity ($S\ m^{-1}$)	Stress	Specific capacitance	Reference
Graphene	0.5	470 kPa(tensile)	160 Fg^{-1}(1 A g^{-1})	Ref. S1
NCG/PPy	8	1.3 MPa(Compression)	-	Ref. S2
Graphene	3.2	-	187 Fg^{-1}(1 A g^{-1})	Ref. S3
PEDOT/GO	73	1.6 MPa(Compression)	174 Fg^{-1}(5 mV s^{-1})	Ref. S4
PVP/PANI	0.05	-	-	Ref. S5
PEDOT/PSS	0.043	600 kPa(tensile)	-	Ref. S6
PEDOT/PSS/Au	0.02	-	-	Ref. S7
PAAm/CMC/PPy	11.0	38 kPa(tensile)	126 Fg^{-1}(0.2 A g^{-1})	This work

Reference

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