## Supplementary Information

Highly Flexible and Mechanically Strong Polyaniline Nanostructure @ Aramid Nanofiber Films for Free-standing Supercapacitor Electrodes

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## 1. Fabrication of ANFs/PANI hybrid films

PANI nanofibers were prepared by rapid polymerization method<sup>1</sup>. Briefly, aniline (1.79 g) and ammonium persulphate (APS) were dissolved in 1 M hydrochloric acid (HCl) of 60 mL, respectively. Then, the APS solution was rapidly poured into the aniline solution, and the mixture was briefly stirred for several minutes. The polymerization reaction was performed at room temperature for 24 h, without stirring or shaking. After polymerization, the mixture was subjected to three cycles of washing-centrifugation (at 10,000 rpm for 15 min). The dark green product was redispersed in water, followed by centrifuging at 3500 rpm for 30 min and collecting the supernatant. The concentration of collecting PANI nanofibers suspension was adjusted to 0.5 mg/mL, which was highly stable in acidic aqueous solution (pH 2.5) for a week. For the preparation of ANFs/PANI hybrid film, desired amount of PANI nanofibers dispersion was slowly added into 25 ml of ANFs/DMSO dispersion (2 mg/mL). The mixture was further stirred vigorously for 2 h to ensure a homogeneous colloidal dispersion before vacuum filtration using nylon membranes with 0.22 µm pore size. The filtered ANFs/PANI hybrid film was rinsed with DI water three times and dried in air. Finally, the ANFs film was peeled off from the filter membrane and dried at 40 °C in vacuum for 3 days. The ANFs/PANI hybrid films with PANI loadings of 5 and 10 wt.% (relative weight ratio of PANI to ANFs) were achieved by controlling the introduced amount of PANI nanofibers dispersion.



Figure S1 (a) Stress-strain curves for ANFs film immersed in 1 M  $HClO_4$  and (b) dependence of ultimate strength on the  $HClO_4$  treatment time for ANFs film.



Figure S2 SEM images (top view) of PANI@ANFs film electrode after 1000 cycles.

PANI loading	Strength	Toughness
(wt.%)	(MPa)	$(MJ/m^3)$
0	255.1 ± 7.1	$30.9 \pm 1.4$
1.6	243.5 ± 5.5	$28.4 \pm 2.3$
4.6	$233.3 \pm 6.1$	$27.8 \pm 1.9$
6.9	$200.6 \pm 8.6$	$21.0 \pm 1.4$
7.7	$192.1 \pm 7.6$	$12.6 \pm 1.1$

Table S1 Mechanical properties for PANI@ANFs film electrodes with different PANI loadings.

## References

1 J. Huang and R. B. Kaner, Angew. Chem. Int. Edit., 2004, 43, 5817-5821.