Supporting Information

Table S1. Conductivity data of CNT-based or graphene-based fibers (Type I), polymer-matrix composite fibers with CNT or/and graphene coatings (Type II), and polymer-matrix composite fibers with CNT or/and graphene fillers (Type III).

Туре	Material	Conductivity	Ref.
		(S m ⁻¹)	
Ι	Graphene fiber	1.11×10^{5}	1
	Graphene fiber	2.3×10^{3}	2
	Graphene fiber	$1.39 imes 10^3$	3
	Poly(vinyl alcohol)-coated graphene fiber	3.5×10^{2}	4
	Poly(vinyl alcohol)-wrapped graphene fiber	9.6×10^{3}	5
	Ag-doped graphene fiber	$9.3 imes 10^4$	6
	CNT fiber	$2.24 imes 10^6$	7
	CNT fiber	1.43×10^{6}	8
	Sodium alginate-modified CNT fiber	3.7×10^{2}	9
	CNT-graphene hybrid fiber	9×10^4	10
	CNT-graphene hybrid fiber	$1.02 imes 10^4$	11
II	Graphene-coated Kevlar fiber	2×10^{3}	12
	Graphene-coated polyurethane/polyester fiber	0.136	13
	Graphene-coated cotton yarn	0.8	14
	CNT-coated Kevlar fiber	6.5×10^{3}	12
	CNT-coated polyester yarn	88.62	15
	CNT-coated cotton yarn	0.4	14
	CNT-coated polycaprolactone fiber	2.85×10^2	16
III	Graphene-polyurethane fiber	0.4	17
	Graphene-polypropylene fiber	3.8	18
	Graphene-poly(ethylene terephthalate) fiber	4×10^{-4}	19
	CNT-polyurethane fiber	0.8	17
	CNT-chitosan fiber	10.7	20
	CNT-polypropylene fiber	2×10^{-4}	21
	CNT-poly(hydroxy ether of bisphenol A) fiber	0.2	22
	CNT-poly(ethylene terephthalate) fiber	0.1	23

Sample	Ato	Atomic concentration (at %)			
	C 1s	N 1s	O 1s		
PI	75.9	3.38	20.72		
GrPI@NAT	81.57	2.89	15.54		
GrPI@AT	80.13	2.22	17.65		

 Table S2. XPS atomic concentration of PI, GrPI@NAT and GrPI@AT yarns.

Sample	Peak area fraction (%)					
	C-C	C-N	C-0	C=O	O=C-NH	O=C-OH
PI	44.88	40.92	6.87	7.33	-	-
GrPI@NAT	36.19	40.85	16.48	6.47	-	-
GrPI@AT	40.53	30.79	13.34	5.77	6.69	2.87

Table S3. C 1s peak area fraction of PI, GrPI@NAT and GrPI@AT yarns.



Figure S1. Comparison on conductivity of CNT-based or graphene-based fibers (Type I), polymer-matrix composite fibers with CNT or/and graphene coatings (Type II), and polymer-matrix composite fibers with CNT or/and graphene fillers (Type III) (data extracted from Table S1).



Figure S2. Cross-sectional SEM images of (a) non-treated and (b) alkali-treated PI yarns.



Figure S3. SEM images of GrPI@AT yarns experiencing (**a-b**) 12, (**c-d**) 7 and (**e-f**) 2 dipping times.



Figure S4. Raman spectra of PI yarn, graphene film and GrPI@AT yarn.



Figure S5. FTIR spectra of PI yarn, GrPI@NAT and GrPI@AT yarns.



Figure S6. (a) Linear mass of graphene on GrPI@AT yarns with increasing the number of dipping GO solution. (b) Weight content of graphene in GrPI@AT yarns with increasing the number of dipping GO solution.



Figure S7. Effect of the conductivity of GrPI@AT yarns on the time of alkali treatment.



Figure S8. Conductivity of GrPI@NAT yarns on the number of times of dipping.



Figure S9. (a) Clear water after the GrPI@AT yarns underwent water wash. (b) Scotch tape test performed on GrPI@AT yarns and clear surface of Scotch tape.



Figure S10. Tensile properties of commercial PI yarn and GrPI@AT yarn.



Figure S11. Electrochemical performance of fiber-shaped supercapacitors using GrPI@AT yarns of different lengths. (**a**) Voltage profile. (**b**) Volumetric capacitance corresponding to different lengths of GrPI@AT yarns. Current density of 90 mA cm⁻³ is used for all cases.



Figure S12. Electrochemical performance of fiber-shaped supercapacitors using GrPI@AT-PANI yarns. (a) Voltage profile. (b) Volumetric capacitance corresponding to different electrodepositing time of PANI. Length of 3 cm and current density of 90 mA cm⁻³ are used for all cases.



Figure S13. Comparison on volumetric specific capacitance between this work and the other supercapacitors based on fibers or textiles.²⁴⁻³⁸

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