Supplementary Information

Highly Twisted Supercoils for Superelastic

Multi-Functional Fibers

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Supplementary Figure 1. Photographs of CNT sheets wrapping process on the stretched $(\varepsilon_{fab} = 400\%)$ spandex core fibre



Supplementary Figure 2. SEM images of the spandex@CNT fibre (a) before, and (b) after twist insertion just before first-coil formation (scale bars = $100 \ \mu m$).



Supplementary Figure 3. Schematic illustrations of (a) supercoiled, (b) coiled, and (c) helical coiled fibres



Supplementary Figure 4. Photograph (upper) and magnified optical images (lower) of four different spots on a 60 cm-long supercoil fibre (scale bar = $500 \ \mu m$).



Supplementary Figure 5. Measured electrical resistance versus length of a 60 cm-long supercoil fibre.



Supplementary Figure 6. SEM image of a SEBS-overcoated spandex@CNT supercoiled

fibre (scale bars = $100 \mu m$)

Supplementary Table 1. Quality factor (Q) comparison of presented supercoiled fibres

with prior-art elastic yarn or fibre conductors

Structures and materials (Ref. No.)	Stretchability	Resistance change	Quality factor
	[ΔL/L₀, %]	[ΔR/R₀, %]	$[\Delta L \cdot R_0 / \Delta R \cdot L_0]$
Spandex@ SEBS/CNT supercoiled fibres (present work)	1,000	4.19	238.8
(a) Buckled CNT@sandwich structured rubber fibre (1)	200	3.7	54
(b) Buckled CNT@SEBS fibre (2)	800		65.1
(c) CNT@nylon coiled fibre (3)	150	1373.7	0.109
(d) Buckled CNT@coiled rubber fibre (4)	400	221.7	1.8
(e) CNT@elastomer polymer fibre (5)	700	450	1.55



Supplementary Figure 7. (a) SEM image (scale bar = $10 \mu m$), and (b) EDX elemental analysis of a MnO₂ coated spandex@CNT supercoiled fibre.



Supplementary Figure 8. Optical image of a supercoil based solid-state supercapacitor at 1000% tensile strain application, which comprises of two parallel, symmetric spandex@ MnO_2/CNT supercoil fires and PVA/LiCl gel electrolyte coating (scale bar = $300\mu m$).

Supplementary Table 2. Comparison of specific capacitance for the supercoil supercapacitor with prior-art yarn or fibre based elastic supercapacitors

Electrode Materials (Ref. No.)	<i>C</i> ^{<i>L</i>} [mF cm ⁻¹]	<i>C_A</i> [mF cm ⁻²]	Stretchability [%]
Spandex@MnO ₂ /CNT supercoiled fibres (this work)	21.7	92.1	1,000
MnO ₂ /CNT@coiled nylon fibres (3)	5.4	40.9	150
Buckled MnO ₂ /CNT@coiled rubber fibres (4)	4.8	22.8	400
Buckled MnO ₂ /CNT@sandwich structured rubber fibres (1)	2.4	11.9	200
PANI/CNT@elastomeric polymer fibres (5)	0.9	50.1	400
MnO ₂ /CNT@spandex, CNT@spandex asymmetric fibres (6)	0.26	27.9	100
CNT/graphene/PANI helical-coil fibres (7)	10.3	273.7	800
PEDOT-S:PSS helical-coil fibre (8)		93.1	400
SEBS@CNT/graphene/MnO ₂ helical-coil fibres (9)		14	850

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