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Separator threads in yarn-shaped supercapacitors to avoid short-circuiting upon length

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Yarn-shaped supercapacitors (YSCs) are becoming promising energy-supply units with decent mechanical flexibility to be integrated into e-textiles in various shapes and locations. However, a robust YSC configuration that can provide long-term and reliable power output, especially after rigorous weaving and knitting processes, as well as all kinds of end uses, is yet to be established. Most YSCs today still suffer from short-circuiting upon length, primarily due to the structure failure of gel electrolyte that also works as the separator. Herein, we report the incorporation of separator threads in a twisted YSC, to withstand repetitive mechanical deformations. Separator threads are wrapped outside of yarn electrodes as a scaffold to accommodate gel electrolyte, while chemistry and wrapping density of these threads are investigated. With processing parameters optimized, we present an YSC configuration that can bear mechanical deformations along almost all directions, leading to reliable power units in woven or knit fabrics.

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INTRODUCTION

Nowadays, wearable-electronics and smart-textiles industries urgently call for flexible energy-storage devices, whereas yarn-shaped supercapacitors (YSCs) and batteries emerged to be plausible solutions in the past decade. YSCs have accumulated growing attention recently, since they carry decent mechanical flexibility to accommodate various fabric structures[1–4], sometimes even sustaining rigorous body movement with minimal addition to fabric mass or volume, especially when compared to their planar counterparts[5]. Meanwhile, YSCs can also provide compatible integration with other textile electronics, such as energy generators, sensors, actuators etc., leading to smart textile systems that can be widely adopted for humans, animals, machines, or even buildings for a variety of applications, such as health monitoring, body injury prevention, environmental sensing, activity tracking, and communications, etc[5–9].

So far, most research on YSCs focuses on improving their electrochemical performance via incorporating high-performance materials (graphene, MXene, MnO_2, conductive polymers etc.) into yarn electrodes, while few discussions are devoted to their structure engineering at the device level[10–15]. Reported YSCs are generally fabricated through coating, paralleling, or twisting two yarn electrodes together in a loosely manner. Coating of active materials onto yarn electrodes results in core-sheath configurations of yarns that often experience ununiform thicknesses and cracks of the coated layers, leading to inconsistency in device performance and sometimes short circuiting[9,16]. Parallel configurations usually result in belt-like devices, which are bulky, easy to fail under mechanical deformations, incompatible with further textile processing, and thus, are mainly adopted for preliminary testing in many reports[5,17–20]. One recent report by Khudiyev et al. went one-step further, where a top-down thermally drawn procedure was adopted to manufacture hundreds of meters long supercapacitor fibers that can be machine weaved[21]. However, it requires sophisticated equipment, flammable organic electrolyte, and bulky polymeric cladding, thereby limiting its volumetric energy density, biosafety in wearable applications, and more importantly long-term stability against electrolyte evaporation and moisture penetration. On the other hand, twisted YSC configurations, where several electrode fibers are twisted together into yarn electrodes and final supercapacitors (twist-yarn supercapacitors, TYCs), are most widely adopted in literature but few reports discussed the details of twisting parameters and their side effects. As a result, it is yet to be developed a reliable and scalable fabrication protocol for TYCs that can indeed satisfy the technological requirements for real application scenarios as mentioned above.

Most TYCs will be subject to repetitive and prolonged mechanical deformations, such as stretching, bending, compression etc., during fabric formation processes (e.g., weaving and knitting) and all kinds of end uses[22]. For instance, in common twisting configurations, the gel electrolyte layer in-between two yarn electrodes is under twisting-induced torsion compression, thereby experiencing localized stress concentration, especially in those thin and stiff yarn electrodes[23–25]. This unique feature requires all the components of TYCs, especially the gel electrolyte that works as both the electrolyte and the separator, to be mechanically robust to maintain their structural integrity, and thus, their energy-storage capability. In our experiments, short circuiting is often observed in TYCs beyond certain length, primarily due to increased chance of damages occurring in the gel electrolyte upon length. Severe safety concerns can arise upon this issue[26], making it a paramount problem to address for TYCs before their practical applications in everyday life. Quite recently, a similar issue was observed in twisted-fiber lithium-ion batteries, wherein a liquid electrolyte was employed and confined within a plastic tube[27]. The authors, He et al., addressed the issue by wrapping a layer of commercial separator film around their fiber electrodes, as a separator layer[27]. However, the adopted commercial film is microporous in nature, which will not function in the same manner as with polymer-gel-based electrolytes, such as PVA/H_2SO_4 or PVA/LiCl gels adopted in most TYCs. The high gel

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viscosity and giant polymeric chains can prevent the gel itself from penetrating through the micropores in those films, which necessitates an alternative solution for TYC systems[28]. In terms of research on strengthening gel electrolyte, toughness of hydrogel electrolytes have been improved by chemical cross-linking, but they usually become too viscous to be continuously coated onto long TYCs[29,30]. For instance, crosslinked hydrogel electrolyte that can resist severe mechanical stimuli has recently been reported by Liu et al.[31]. However, this electrolyte does not provide enough fluidity to be incorporated into continuous TYCs. Instead of pure chemical cross-linking, fiber-reinforced hydrogel is a more practical strategy to improve their overall mechanical performance while maintaining structure integrity[32–35]. Li et al.[32] also found that the load-bearing and anti-wearing capabilities of hydrogel can be largely improved via fiber reinforcement.

Inspired by the idea of fiber-reinforced hydrogels, we introduce insulating fibers as separator threads into our TYCs to ensure the structure integrity of gel electrolyte, protecting the TYCs from short circuiting in various load-bearing activities. Though non-conductive fibers have been used as spacer to separate two parallel short fiber electrodes that are confined in plastic tubes with liquid electrolyte[36,37], the continuous fabrication and detailed structure engineering of separator threads in gel electrolyte based TYCs are yet studied. Here, a scalable fabrication process has been developed in our lab to incorporate the separator threads at predetermined structure parameters, wherein continuous TYCs can be easily manufactured. The resulted YSCs are flexible, mechanically robust, and electrochemically reliable, enabling them to survive various mechanical stresses, such as bending, stretching, and compression in practical applications.

RESULTS AND DISCUSSION

Optimization of the wrapping density of separator threads

To clearly demonstrate the effect of separator threads in TYCs, we adopt a simple TYC structure as the exemplar, where activated carbon (AC) is chosen as the active electrode material, commercial Carbon fiber yarns (CYs, 1k) are employed as current collectors, and PVA/LiCl system is used as the gel electrolyte, mainly due to its hygroscopic nature[38] that ensures the long-term performance stability of resulted TYCs (Supplementary Fig. 1). Our electrode yarns are prepared via loading certain amount of AC into CYs (AC-CYs). With 10 wt.% of loading (~110 μm in diameter), AC-CYs yarn electrode offers a capacitance density of 12.2 mF cm^{-2} (0.1 mF cm^{-1} @20 mV s^{-1}), and when the loading increases to 50 wt.% (~160 μm in diameter), its capacitance density increases to 233.7 mF cm^{-2} (3.7 mF cm^{-1} @20 mV s^{-1}) at 10-cm length (Supplementary Fig. 2), outperforming many previously reported supercapacitor fibers or yarns (Supplementary Table 1)[39–45]. A TYC is usually fabricated via twisting two gel-electrolyte-coated yarn electrodes at certain level of twisting (expressed as twists per cm, i.e. tpc) (Supplementary Fig. 3a)[46,47]. As shown in Supplementary Fig. 3b left, for a 20-cm TYC that is **manually** twisted at 3 tpc, the electrolyte layer (~150 μm thick, highlighted in red) can remain intact along the yarn length, and the YSC offers reasonable capacitance and equivalent series resistance (ESR) (Supplementary Fig. 3c, d). However, when it is twisted by a customized thread twisting machine (from *Domanoff workshop*), the coated gel electrolyte is partially stripped off from the yarn electrode surfaces due to abrasion (Supplementary Fig. 3b, middle). When the twisting level further increases to 5 tpc, the electrolyte layer becomes thinner in between the two electrodes, resulting in incomplete physical barrier as a separator (Supplementary Fig. 3b, right). Consequently, the TYC is experiencing short-circuiting in both machine-assembled cases (tpc 3&5, red and blue curves in Supplementary Fig. 3c, d). Therefore, it is crucial to protect the gel electrolyte layer in TYCs for them to perform reliably. For this purpose, separator threads are introduced into our TYCs, while the chemical ingredients of these threads should be inert to salt, acid, or alkali solutions. For example, cotton yarns cannot survive strong acid due to its acid-catalyzed hydrolysis, thus are excluded in acidic electrolyte despite their natural abundance and decent hydrophilicity. In addition, separator threads should have decent affinity to the gel electrolyte, small in diameter to minimize the separation distance for electrolyte ion diffusion.

In our experiments, a type of nylon multifilament threads (tex 16, ~110 μm in thickness) is chosen as the separator threads since it's light, flexible, and cost-effective, satisfying all the criteria mentioned above. The nylon threads are first wrapped outside of the electrode yarns via a yarn twister, where the electrode yarn (AC-CY) is fixed as the core, and a few nylon threads are wrapped as the sheath to produce a core-sheath structure (Fig. 1a).

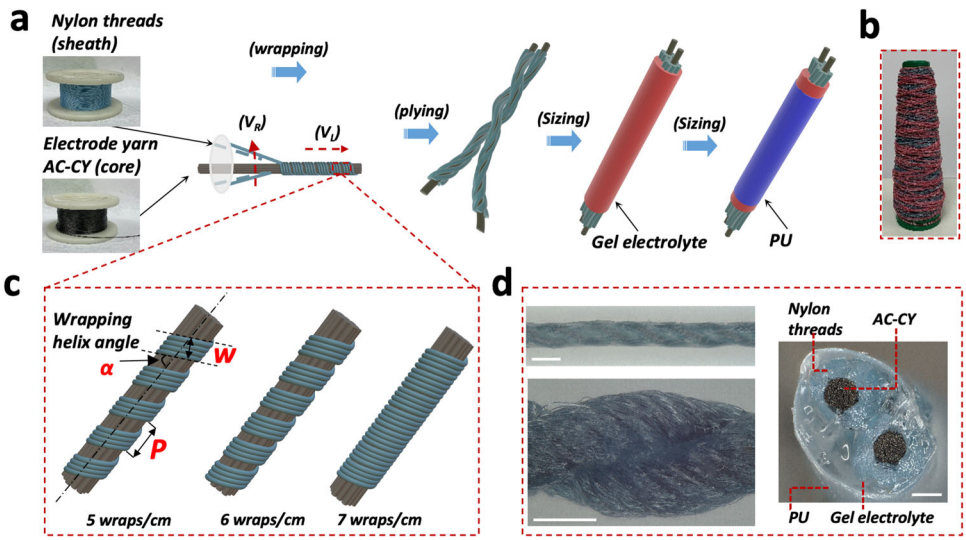

Fig. 1 Depiction of our TYCs with schematics, photos, and optical microscopic images. a Schematic diagram of the fabrication process of TYCs with separator threads incorporated; **b** Tens of meters long TYCs on a spool (the color comes from different nylon separator threads incorporated); **c** Schematic illustration the wrapping density of separator threads in AC-CYs; **d** Optical microscopic images showing the side and cross-sectional views of our TYCs (scale bar in left top: 1 mm; left bottom: 500 μm; right: 250 μm).

Sheath nylon threads are distributed evenly with certain wrapping density, which can be tuned by controlling the yarn take-up speed (V_L) and sheath yarns rotating speed (V_R) (Fig. 1a). Afterwards, two core-sheath yarn electrodes are plied together into a two-ply yarn (Fig. 1a) with the same yarn twister. Subsequently, PVA/LiCl gel electrolyte is infiltrated into the TYCs (details in Supplementary Fig. 4). The continuous electrolyte coating process can be conducted with a sizing machine, which is commonly used in textile industry for sizing of warp yarns[48]. Finally, to avoid the surface stickiness of the PVA/LiCl gel electrolyte and to protect YSCs from contamination, TYCs are coated with a protective layer of polyurethane (PU). With these consecutive steps, continuous TYCs can be fabricated in hundreds of meters (Fig. 1b). The wrapping density of separator threads and the twisting level between the two yarn electrodes are the major structural parameters dictating the performance of the resulting TYCs.

Cotton and Polyethylene Terephthalate (PET) threads in similar thicknesses have also been employed in our devices as separator threads (Supplementary Fig. 5a). Both lead to comparable electrochemical performance as the nylon threads according to the same fabrication protocol (Supplementary Fig. 5b, c), although cotton absorbs more gel electrolyte and PET catches less as compared to nylon. Therefore, the chemistry of the separator threads poses minimum impact on TYC performance, as long as these threads are chemically inert in the electrolyte, applied at appropriate wrapping density, and carry good affinity to the electrolyte. To further investigate the gravimetric and volumetric performance of TYCs, we stick to nylon threads due to their lower mass addition to the devices (Supplementary Table 2).

Separator threads are introduced to maintain the structure integrity of gel electrolyte layer, preventing TYCs from short-circuiting. This desired effect is largely governed by how separator threads are wrapped outside of each yarn electrode. In our experiment, four nylon threads as sheath components move as a group with a width (W) of ~1 mm, going helically around the core AC-CY yarn (Fig. 1c). This group of separator threads can vary in wrapping helix angle (α) and thus wrapping helix pitch (P) (Supplementary Fig. 6). Wrapping density can be expressed in the number of wraps per unit length of the core-sheath yarn (e.g. wraps/cm). As wrapping density increases, α increases, resulting in decreased P and D, the uncovered distance in between two wrapped area, and thus less exposed electrode surfaces (Supplementary Fig. 6). Based on the thickness of AC-CY yarns (~150 µm) and the wrapping width (W) of nylon threads (~1 mm), we compare TYC performances at three different wrapping densities: 5 wraps/cm, 6 wraps/cm, and 7 wraps/cm, respectively.

As illustrated in Fig. 1c, the uncovered distances in each pitch of the separator threads are *ca*. 1 mm at 5 wraps/cm, *ca*. 200 µm at 6 wraps/cm, and minimum at 7 wraps/cm (almost entire surface covered), respectively. Subsequently, two such AC-CY electrode yarns will have to be twisted (also named as 'plying' by textile engineers) together to form the final TYC, while this additional layer of plying (characterized by plying tpc) is inevitably imposing extra compression onto both electrodes. As a result, TYCs at 5 wraps/cm still experience some short-circuiting as the plying tpc of two electrode yarns increases from 2 to 6 (Supplementary Fig. 7). This indicates that the two electrode yarns can still contact each other across the separator threads upon pressure, especially when they are conformed into a more compact structure under a higher plying tpc. Interestingly, no short-circuiting is observed in TYCs with separator-threads wrapping at 6 and 7 wraps/cm, even at higher plying tpc of electrode yarns. In addition, higher wrapping density, i.e. 7 wraps/cm, can result in slightly lower ESR (Supplementary Fig. 8), which can be attributed to the bigger helix angle (α) of the wrapped threads. The lateral force generated from the sheath threads' stress on the core yarns can increase monotonically with the increase in helix angle[49,50]. As a result, the core yarn can be contracted more closely, which is beneficial for the contact between AC particles and CYs, and thus, reducing the interfacial resistance accordingly. As shown in Fig. 1d, the TYC at 7 wraps/cm exhibit a compact and uniform structure along the longitudinal direction. The nylon separator threads are tightly and evenly wrapped around the AC-CY electrodes, fully filled with gel electrolyte across their entire thickness (Fig. 1d). Therefore, we adopt 7 wraps/cm as the wrapping density to fabricate TYCs for all the following study.

Effect of plying on TYC electrochemical and mechanical performance

Within a two-ply yarn structure of our TYCs, plying level (in tpc) decides the compression pressure and actual length of the two plied yarns (Fig. 1a), thus can lead to different electrochemical performances of TYCs. For example, higher tpc will result in longer yarn electrode and more active materials within the unit length of TYC (Supplementary Table 3), providing higher linear capacitance accordingly. Plying level also poses great impact on the mechanical properties of TYCs, affecting their compatibility with machine weaving and knitting processes.

Based on the thickness of nylon separator threads, we applied three levels of plying in our TYCs (Fig. 2a), including a low level of 2 tpc (TYC-2), a medium level of 4 tpc (TYC-4) and a high level of 6 tpc (TYC-6). As the tpc further increases to 7, the electrode yarns are snarling up due to over twist (Supplementary Fig. 9). As the tpc increases from 2 to 6, the length of electrode yarns in a 10-cm-long TYC increases from 10.3 cm at 2 tpc, to 10.8 cm at 4 tpc, and to 11.6 cm at 6 tpc (Supplementary Table 3). As a result, the linear capacitance of TYCs (50 cm in length) increases (Fig. 2b) from 2.2 mF cm^{-1} for TYC-2, to 2.6 mF cm^{-1} for TYC-4, and to 3.0 mF cm^{-1} for TYC-6, respectively. Their ESRs remain comparable though, despite the length discrepancy (Fig. 2c). In terms of gravimetric performance, with increasing tpc, the mass of TYCs before electrolyte coating (Supplementary Table 3) increases proportionally due to the longer electrode yarns in unit length of TYCs. However, after electrolyte coating (same coating process for all three TYCs), TYC-2 becomes the heaviest, since its loose structure can accommodate more gel electrolyte. TYC-4 and YSC-6 are relatively compact, exhibiting less extra room for electrolyte. As a result, TYC-6 presents the highest gravimetric capacitance of 0.8 mF g^{-1}, 16% and 52% higher than that of TYC-4 and TYC-2, respectively. Therefore, TYC-6 provides both the highest linear and gravimetric capacitances in our experiments.

The tensile behaviors of TYCs at different plying levels are also evaluated as shown in Fig. 2d. There are four components in our final TYCs, including two AC-CY electrode yarns, nylon separator threads, PVA/LiCl gel electrolyte and a surface layer of PU coating. When a TYC is stretched, all the components are under stress simultaneously. But each component is experiencing breakage separately due to their different mechanical properties. Comparing to other components, the two AC-CY electrode yarns with the lowest deformation capability and relative short length will break first, which is reflected as the first peak on the stress-strain curve (Fig. 2d), indicating the elasticity of the electrode yarns and the proper function of the TYCs upon stress. As shown in Fig. 2d, TYC-2 possess the tensile strength of ~300 MPa for the first peak, higher than those of TYC-4 and TYC-6, whose tensile strengths are both ~200 MPa. On the contrary, in terms of the elongation (when carbon fibers start to break), the strain of TYC-6 is the highest at ~15%, followed by TYC-4 at ~5%, and TYC-2 at ~2%. Therefore, plying AC-CYs into this composite structure circumvents CYs' brittleness and low elongation nature (<2%), producing TYCs with tensile strain (>4%) that is suitable for conventional weaving. In addition, the increase in plying can lead to increases in stiffness and fiber compactness in TYCs, making them customizable to adapt to different fabrication processes and end-use applications.

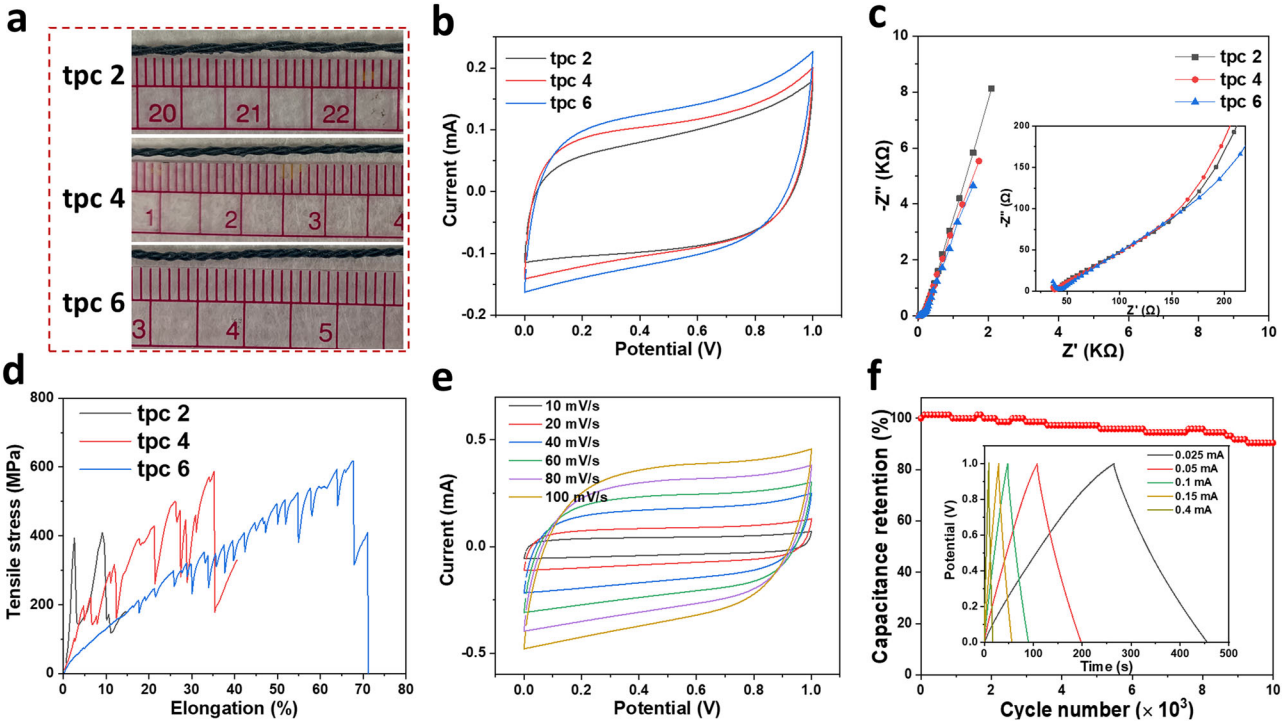

Fig. 2 Electrochemical and mechanical performances of TYCs at different plying levels. a Photos of TYCs with different ply-twist degrees, including tpc 2, tpc 4, and tpc 6; **b**, **c** CV curves (40 mV s^{-1}) (**b**) and Nyquist plots (**c**) of TYCs (50 cm) with different ply-twist degrees; **d** Typical stress-strain curves of TYCs with different ply-twist degrees; **e**, **f** CV curves at different scan rates (**e**) and capacitance retention upon 10,000 charge-discharge cycles, with the inset figure showing the GCD curves at different current densities (**f**) of a 50-cm-long TYC with tpc 4.

To better understand the electrochemical performance of the TYCs, we show the cyclic voltammogram (CV) and galvanostatic charge-discharge (GCD) curves of a 50-cm long YSC (at tpc 6). As indicated by the CV curves at different scan rates in Fig. 2e, the YSC maintains quasi-rectangular shape up to the scan rate of 100 mV s^{-1}. The GCD curves exhibit nearly symmetrical triangular shape, indicating good coulombic efficiency of ~94% (Fig. 2f inset). The capacitance of TYCs is retained 90% over 10,000 cycles of charge-discharge at current density of 0.05 mA (Fig. 2f).

The stability of TYCs against mechanical deformation

When YSCs are being processed into fabrics for real applications, they are often subjected to various mechanical distress, including bending, stretching, and compression. To demonstrate the bending flexibility of our TYCs, a 50-cm-long TYC (at 7 wraps/cm and 6 tpc) was densely wrapped on tubes in different diameters, 5 mm, 3 mm, and 1.5 mm, respectively (Fig. 3a). The TYC wrapped on a thinner tube is under higher degree of bending. The smaller the diameter, the greater the required flexibility of the yarn. Flexibility is an important characteristic for knitting yarn, which enables yarns to be smoothly transformed into loops that compose a knit structure. At all the three levels of bending, the TYC exhibits almost the same electrochemical performance as that of the original state (straight, Fig. 3b, c), indicating good bending stability.

Other than bending, TYCs will also bear various loads along both the longitudinal and transverse directions. Herein, we evaluate the stability of our TYCs against stretching and compression, respectively. In terms of stretching, the TYC is used to lift a 1 kg weight (equal to 156 MPa stress applied on TYCs) several times (Fig. 3d). Specifically, the TYC is tied with a cotton yarn that is fixed on the weight. As shown in Fig. 3e, f, after lifting the weight up for 100 times, the TYC can still maintain its original electrochemical performance. Regarding transverse compression, a TYC is put underneath a 1 kg weight (~ 580 Pa) for up to ~120 h (Fig. 3g). The TYC can well maintain its intact structure and thus its electrochemical performance (Fig. 3h, i). In contrast, an originally well-functioned TYC without separator threads (at tpc 3) can easily get shorted and lost their functionality upon stretching and compression (Supplementary Fig. 10). With separator threads incorporated as scaffold for gel electrolyte, TYCs exhibit reliable load-bearing capability along both longitudinal and transverse directions, holding great promise for real-life applications.

The connection of TYCs for real applications

Other than PVA/LiCl based electrolyte, TYC with another type of electrolyte can also be fabricated via our established protocols, as long as compatible separator threads are selected. For instance, a well-functioning TYC with PVA/H_2SO_4 based electrolyte can be assembled with PET threads (Supplementary Fig. 11). In addition, to enlarge the capacitance in the unit length of TYCs, two or more electrode yarns (AC-CYs) can be firstly twisted into a multi-plied yarn, then wrapped with separator threads. The wrapping density of separator threads can be adjusted based on the thickness of the multi-plied electrode yarn. The length normalized capacitance of a 'two-ply AC-CY' TYC increases to 3.9 mF cm^{-1} from 1.9 mF cm^{-1} of a 'single AC-CY' TYC, and the volumetric capacitance increases to 1.2 mF cm^{-3} from 0.74 mF cm^{-3}, while there is only a 30% increase in device volume (Supplementary Fig. 12).

To meet the voltage requirement in real applications, TYCs can be easily connected in series to expand the voltage window. For example, four serially connected TYCs can readily expand its voltage window up to 3.2 V (Fig. 4a) to power up a light-emitting diode (LED). As shown in Fig. 4b, when four 10-cm YSCs are connected in series, they can provide adequate energy to light up a red LED.

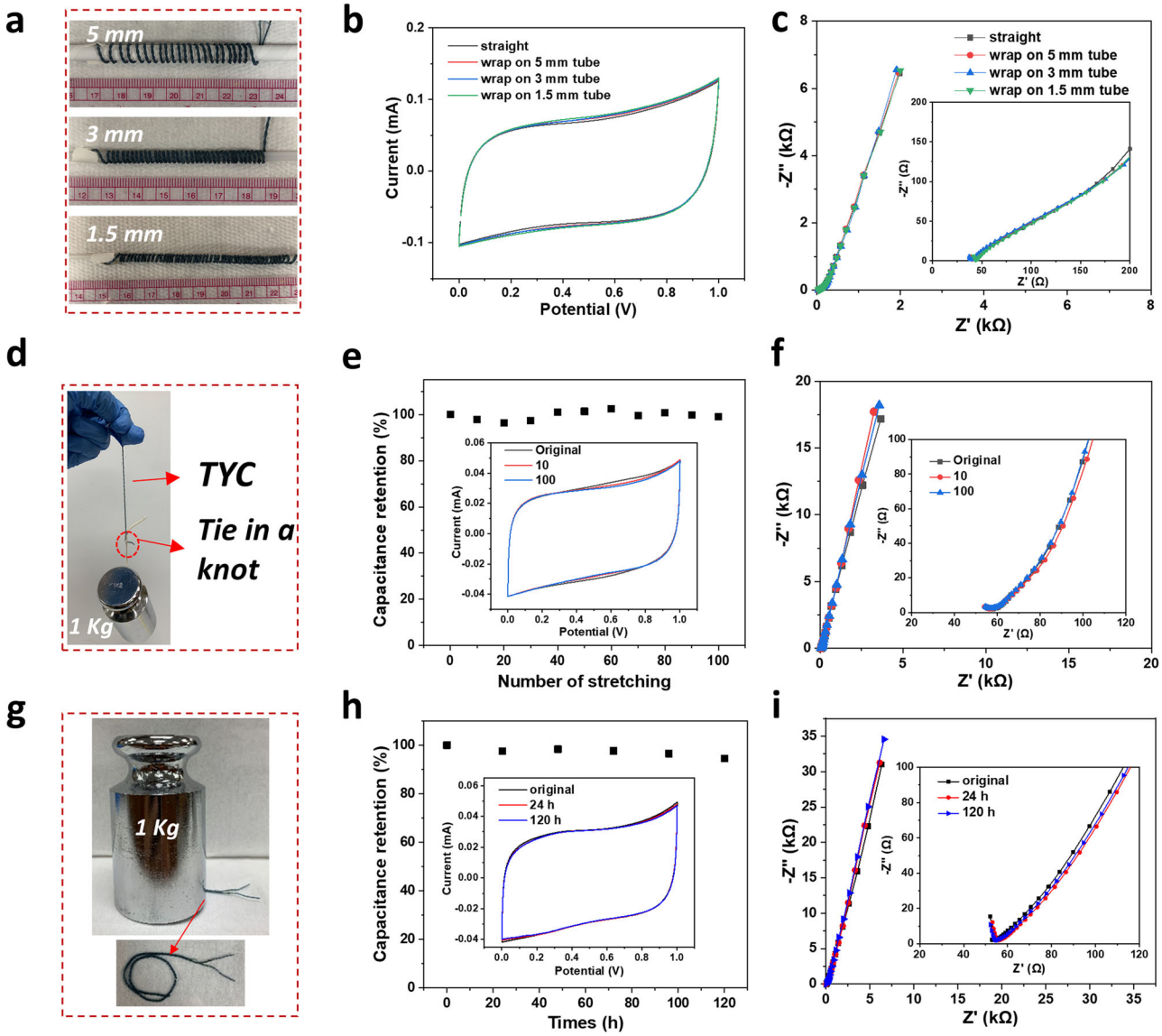

Fig. 3 Stability of TYCs against mechanical deformation. a Photos of TYCs wrapped on tubes in diameters of 5 mm, 3 mm, and 1.5 mm, respectively; **b**, **c** CV curves (40 mV s^{-1}) (**b**) and Nyquist plots (1 M–0.01 Hz), with the inset figure zoomed in at the high-frequency domain of the plots (**c**) of TYCs in straight state and wrapped on tubes; **d** Photo of a TYC lifting a 1 kg weight; **e** capacitance retention of TYC upon numbers of stretching, with the inset figure showing the CV curves (40 mV s^{-1}); **f** Nyquist plots (1 M–0.01 Hz) of the original, 10th and 100th stretching of the TYC, with the inset figure zoomed in at the high-frequency domain of the plots; **g** Photos of an YSC under compression of a 1 kg weight; **h** capacitance retention of TYC upon hours of compression, with an inset figure showing CV curves (40 mV s^{-1}); **i** Nyquist plots (1 M–0.01 Hz) of the TYC under compression for different time periods, with the inset highlighting the high-frequency domain of the plots.

A side benefit of separator threads is that their color can be used to distinguish the two electrode yarns in TYCs. As illustrated in Fig. 4c, one electrode is wrapped with dark blue threads, and another is with orange threads. The colorful TYCs also offer versatile design flexibility and improve the aesthetic of resulting fabrics. A well-functioned TYC (1.6 m) can be readily fabricated with no short-circuiting observed (Supplementary Fig. 13). The flexible and strong TYCs can be incorporated into both woven structure (Fig. 4d) and knit structure (Fig. 4e), while their original electrochemical performance is well maintained (Supplementary Figs. 14 and 15).

In this work, we have demonstrated the necessity of separator threads in twisted-yarn supercapacitors (TYCs, and perhaps in coated and parallel configurations as well), for them to maintain reasonable electrochemical output against repetitive mechanical deformations such as bending, stretching, and compression, etc. The chemistry, wrapping density, and helix angle of the separator threads are compared and optimized in our system, wherein activated-carbon coated carbon-fiber yarns are adopted as electrodes, and PVA-based LiCl gel is employed as the electrolyte. We now, with high confidence, present a reliable and scalable fabrication protocol for TYCs that can bear mechanical deformations along different directions and function as reliable power sources in woven or knit fabrics.

METHODS

Materials for twisted-yarn supercapacitors (TYCs)

The activated carbon (AC, Kuraray Co., LTD), carbon black (IMERYS) were used as received without further treatment. 1k carbon fiber yarns were

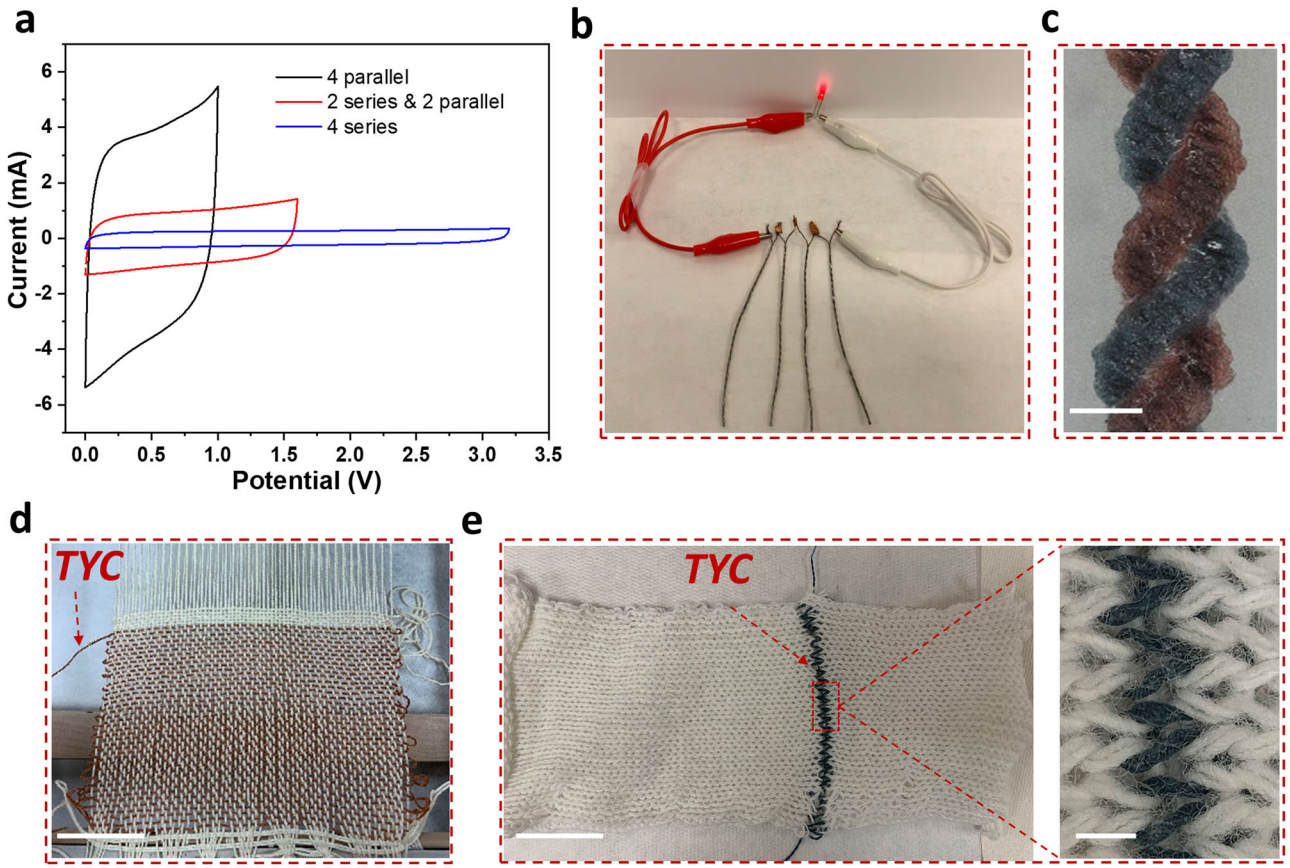

Fig. 4 Demonstration of TYCs for real applications. a CV curves (40 mV s^{-1}) of four TYCs in different connecting strategies; **b** Photo of four serially connected TYCs lighting an LED; **c** Photo of a TYC with each electrode wrapped with different color of nylon threads (Scale bar: 1 mm); **d** Photo a woven fabric with TYCs (orange) incorporated as weft yarns (Scale bar: 10 cm); **e** Photo of a knitted fabric with a one-meter long TYC (dark blue) incorporated (Scale bar in left: 10 cm; right: 2 cm).

purchased from *CST The Composite Store*. Poly (vinyl alcohol) (PVA; Mw 146,000–186000, 99 +% hydrolyzed), and glutaraldehyde solution (25% in H_2O) were purchased from Sigma-Aldrich. Lithium Chloride (LiCl), and orthophosphoric acid (H_3PO_4; ≥ 85%) were from Fisher Scientific and VWR, respectively. Nylon thread was purchased from *The Thread Exchange, Inc*. Polyurethane (Desmocoll® 530/3) was from Covestro.

Fabrication of yarn electrodes and TYCs

AC-CY preparation: AC particles are mixed with carbon black, PVA (5 *wt*.%) and 1 M H_3PO_4 in deionized water (DI) H_2O. The ratio of AC:carbon black: PVA was 90:5:5 by weight. The mixture then was loaded into 1k CY via the sizing machine.

Nylon threads were wrapped outside of AC-CY at a fixed wrapping density in 'Z' direction. Then, two wrapped AC-CY yarns were plied at certain twist level in the opposite 'S' direction to form a balanced and tight two-ply yarn. Afterwards, PVA/LiCl based electrolyte was coated and penetrated into the two-ply yarns to form the functional TYC. Finally, to coat a protective layer of PU outside of the TYC, the PU was dissolved in acetone at the concentration of 1 mg mL^{-1}, then coated onto TYC continuously with acetone evaporated later at room temperature (Supplementary Fig. 16).

Structure characterizations

The structure and morphology of TYCs were characterized by VHX-7000 digital Microscope. The tensile test was performed on the Q-test system. The test conditions were 10 mm min^{-1} extension rate and 10 cm gauge distance.

Electrochemical performance characterizations

Electrochemical performance, including cyclic voltammetry (CV), galvanostatic charge/discharge (GCD) measurements, and electrochemical impedance spectroscopy (EIS) analysis were conducted using electrochemical workstation (Autolab, Metrohm, USA). Two yarn electrodes at the same end of the TYCs were directly connected to the electrochemical workstation for electrochemical measurements. The capacitance can be calculated from CV curves according to the equation: $C = (1/2v) \times \oint IdU/\triangle U$, where v is the scan rate, I is the instantaneous current, $\triangle U$ is the voltage range. C could also be calculated from GCD test: $C = t \times I/(\triangle U - U_{drop})$, where t is the discharge time, I is the discharge current, $\triangle U$ is the potential window, U_{drop} is the voltage drop in the discharge curve. The energy density (E) and power density (P) of the TYC can be obtained from $E = C\_device \times U\hat{}2/7200$, $P = (\triangle U - U_{drop})^2/4 * ESR$. ESR is the electrochemical series resistance of the device that can be calculated from the formula $ESR = U_{drop}/2I$. The areal, volumetric and length capacitances were calculated from single yarn electrode. The area and volume of yarn electrodes were calculated using the following equations: Area: $S = L\pi D$, Volume: $V = L\pi(D/2)^2$, where L and D are the length and average diameter of yarn electrode, respectively. The areal and volumetric energy density and power density were calculated based on the area of the entire TYC device.

Weaving and Knitting

The woven fabric was achieved by a hand-loom (Ashford rigid heddle loom with reed in size 7.5 dents/inch) and worsted weight cotton yarns were used as warp yarns. The knitting fabric was achieved by a circular sock knitting machine (equipped with gauge 3 needles).

DATA AVAILABILITY

The data that support the finding of this study are available from the corresponding author upon reasonable request.

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AUTHOR CONTRIBUTIONS

N.H., F.Z., and W.G. conceived this work. N.H., J.S., and J.L. performed the experiments. N.H. and W.G. wrote the manuscript with inputs from all coauthors. All authors read and commented on the manuscript.

COMPETING INTERESTS

The authors declare no competing interests.

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