

Energy harvesting and storage in 1D devices

Hao Sun*, Ye Zhang*, Jing Zhang, Xuemei Sun and Huisheng Peng

Abstract | Power systems and electronic devices that are bulky and rigid are not practical for use in wearable applications that require flexibility and breathability. To address this, a range of 1D energy harvesting and storage devices have been fabricated that show promise for such applications compared with their 2D and 3D counterparts. These 1D devices are based on fibres that are flexible and can accommodate deformation, for example, by twisting and stretching. The fibres can be woven into textiles and fabrics that breathe freely or can be integrated into different materials that fit the curved surface of the human body. In this Review, the development of fibre-based energy harvesting and storage devices is presented, focusing on dye-sensitized solar cells, lithium-ion batteries, supercapacitors and their integrated devices. An emphasis is placed on the interface between the active materials and the electrodes or electrolyte in the 1D devices. The differing properties of these interfaces compared with those in 2D and 3D devices are derived from the curved surface and long charge transport path in 1D electrodes.

Over the past decades, electronic devices have, in general, become more powerful and smaller in size^{1–3}. For example, the first general-purpose electronic computer (ENIAC) weighed over 27 tonnes, occupied 167 m² and operated at 5,000 cycles per second⁴, but nowadays, portable computers fit in our briefcases and operate at billions of cycles per second. In addition, electronic products such as Apple Watch, Google Glass and sport wristbands enable us to communicate with others or detect the status of our health. As a result of these trends in size and human behaviour, electronic devices are now required to work in close contact with the human body. However, the development of these devices is hindered by obsolete power systems that are bulky and rigid. In particular, conventional energy harvesting and storage devices, including silicon-based solar cells, dielectric capacitors, lead-acid and lithium-ion batteries, fail to meet the flexibility required for wearable electronics⁵. Hence, it is crucial to develop energy harvesting and storage devices with competitive performances and durability, as well as the flexibility that allows their fabrication and function in wearable situations^{6,7}.

One-dimensional energy harvesting and storage devices reported in the past decade have begun to meet the requirements of wearable electronics. These devices typically take the form of flexible fibres with diameters ranging from tens to hundreds of micrometres, which can accommodate complex deformations, such as twisting and stretching on irregular substrates^{8,9}. In addition, it is possible to weave such fibre-based devices into

breathable textiles or integrate them to form deformable structures^{10–12}.

The fabrication of 1D energy harvesting and storage devices can be achieved using two main structures — namely, **coaxial** and **twisted structures** (FIG. 1a). A coaxial structure has a core–shell architecture with a fibre electrode core, an electrode outer shell and an active material sandwiched in-between^{13–15}. The fabrication of coaxial devices involves coating an active material on a fibre followed by the preparation of an outer electrode layer. This process is simple and can be easily scaled up. However, it remains challenging to coat thin, uniform films on a highly curved fibre, and the coated film easily breaks under deformation. It is also difficult for the active material or thin electrode layer to simultaneously meet the requirements of high optical transmittance, high tensile strength and low electrical resistance¹⁶. For example, the outer shell of a coaxial dye-sensitized solar cell (DSSC) is expected to be as thin as possible to achieve high light transmission for high power conversion efficiency. However, a thin outer electrode layer typically shows larger electrical resistance, which decreases the power conversion efficiency. As a consequence, a balance between transmittance and resistance is sought after and an optimum thickness is used. For example, in a coaxial DSSC with dye-adsorbed titania-modified titanium wire as the inner electrode and aligned multi-walled carbon nanotube (MWCNT) sheets as the outer electrode, an **optimum MWCNT thickness of 20 nm** has been revealed¹⁷ and is now widely used for fibre-based solar cells^{18,19}.

State Key Laboratory of Molecular Engineering of Polymers, Department of Macromolecular Science, and Laboratory of Advanced Materials, Fudan University, Shanghai 200438, China.

*These authors contributed equally to this work.
Correspondence to X.S. and H.P.
sunxm@fudan.edu.cn; penghs@fudan.edu.cn

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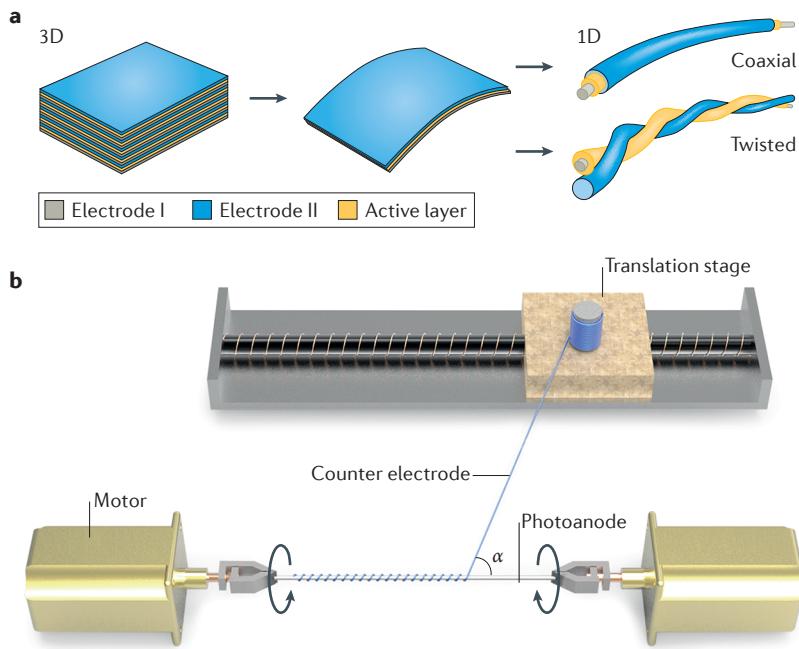


Figure 1 | Evolution of energy harvesting and storage devices from 3D to 1D.

a The 1D devices are typically either coaxial or twisted structures. **b** A rotation–translation setup for the production of fibre-based dye-sensitized solar cells with a twisted structure. A counter electrode fibre is fixed on a translation stage and continuously wound around a photoanode fibre stabilized with a motor at both ends. The twisting angle (α) is controlled by the initial angle between the photoanode and counter electrode and the twisting and releasing speeds of the counter electrode, which are realized by balancing the rotating speed of the motors and the velocity of the translation stage.

In a twisted structure, two fibre electrodes are incorporated with active materials and then wound together at a certain twisting angle^{20–22}. The twisting process generally uses a rotation–translation setup (FIG. 1b). Typically, a photoanode is stabilized with a motor at each end, and a counter electrode is fixed to a separate translation stage. The counter electrode is then attached onto the photoanode at a certain initial angle, and then the two motors and translation stage begin to move simultaneously. The twisting angle ($\alpha = 60^\circ$ in FIG. 1b) can be achieved by setting up an initial angle between the photoanode and counter electrode and maintaining the same twisting and releasing speeds of the counter electrode, which is realized by balancing the rotating speeds of the motors and the velocity of the translation stage²³. When the twisting angle is zero, the two fibre electrodes form a parallel arrangement²⁴. This twisting process can be automated and allows continuous production; however, this is more difficult to achieve for coaxial structures largely because of the absence of a thin electrode shell that is present in the twisted structures. Although the twisting process may be considered an added complication, the integrity of devices fabricated without twisting of the component fibre electrodes is often relatively low, and hence their practical application is hindered.

From a materials perspective, nanomaterials attract the most interest for use in fibre-based energy harvesting and storage devices because of their large surface areas and remarkable mechanical, electronic and thermal

properties²⁵. The resulting interfaces between active materials and the electrolyte, between active materials and electrodes, and between neighbouring building blocks of the electrode differ in 1D and 2D configurations. In this Review, the recent advances of 1D energy harvesting and storage devices, with an emphasis on these interfaces, are outlined. The integration and weaving of the fibres are also described to highlight the challenges for future development in this field. In addition, the fabrication of multifunctionalized devices that incorporate additional properties such as self-healing²⁶, colour change²⁷ and shape memory²⁸ are outlined. The integration of 1D energy harvesting and storage devices is generally achieved by consecutively coating energy harvesting and storage materials on a fibre²⁹ (to form an in-series architecture) or sequentially depositing them from inside out³⁰ (to form a core–shell architecture). The weaving of these 1D devices by well-developed textile technology can obtain flexible materials for wearable applications^{10,12}.

Timeline of developments

In 2002, 1D energy harvesting devices emerged following the coating of photoactive materials onto a metal wire to yield 1D coaxial DSSCs¹³ (FIG. 2). This was followed some years later by the fabrication of 1D polymer solar cells by coaxially coating photoactive materials on optical fibres¹⁴. In 2008, 1D DSSCs were produced by twisting together two electrodes composed of metal wire²⁰. In more recent years, the photovoltaic performance of 1D DSSCs has been optimized by introducing electrodes comprising lightweight and flexible aligned MWCNTs²¹. In 2014, to further enhance the power conversion efficiency, 1D perovskite solar cells were produced to form all-solid-state candidates¹⁸. In addition, other energy harvesting devices — namely, 1D piezoelectric and triboelectric nanogenerators — were developed in 2008 and 2014, respectively^{31,32}.

In terms of energy storage devices, 1D supercapacitors with a twisted structure were proposed in 2003 (REF. 22), and ten years later, coaxial devices^{15,33} and stretchable 1D supercapacitors were reported³³. At a similar time, flexible 1D lithium-ion batteries were produced³⁴, and the electrochemical performances have been further enhanced by the emergence of 1D zinc-air batteries³⁵, lithium–sulfur batteries³⁶, lithium–air batteries³⁷ and aluminium–air batteries³⁸. The integration of 1D energy harvesting and storage devices was realized by the incorporation of a DSSC with a supercapacitor on a titanium dioxide-deposited titanium wire in 2012 (REF. 29). A multifunctional 1D energy harvesting device that could be remotely operated by a magnetic field appeared in 2013 (REF. 39).

Using 1D structures in electronic devices raises concerns that were not relevant for 2D and 3D structures. For example, the conventional strategy of using an insulating separation membrane is technically difficult for a thin-fibre-based device. As a result, a short circuit is more likely to occur in a fibre-based power system based on liquid electrolytes owing to the contact of two fibre electrodes under deformation. To this end, gel or solid electrolytes are typically used to confine the fibre electrodes.

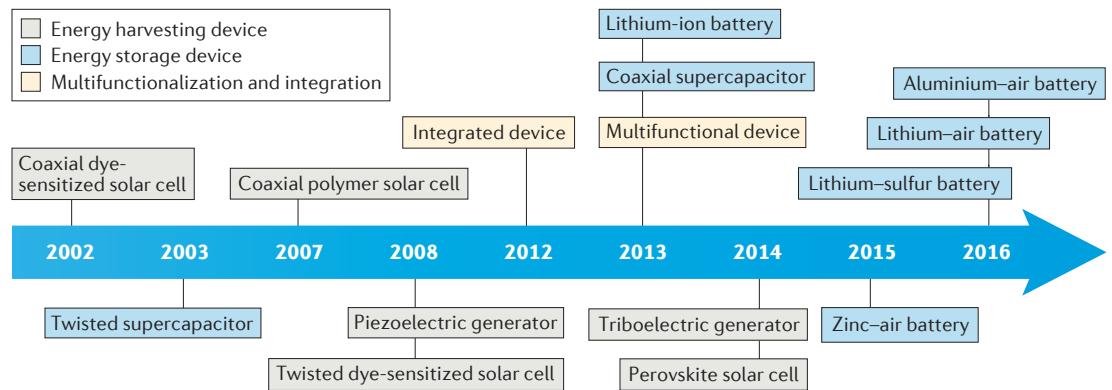


Figure 2 | Timeline of developments in 1D energy harvesting and storage. Energy harvesting devices include solar cells and nanogenerators, and energy storage devices include supercapacitors and batteries. **Multifunctionalization** involves energy harvesting and storage devices that are incorporated with additional functionalities such as self-healing²⁶, changes in colour²⁷ and shape memory properties²⁸. The integration of 1D energy harvesting and storage devices into one system is generally achieved by consecutively coating energy harvesting and storage materials along the length direction of a fibre²⁹, or sequentially depositing the energy storage part as the core and the energy harvesting part as the shell³⁰.

Also, materials that are electrically insulating but conduct ions have been coated on the surface of the fibre electrodes to prevent them from contacting each other⁹. Recently, using an intricate laser method, the opposing sides of a graphene oxide fibre were reduced to form graphene ribbons serving as two electrodes of a fibre-based supercapacitor. The resulting 1D device was shown to maintain over 75% of the initial capacitance after 160 bending cycles and no short circuits were detected⁴¹. It is important to maintain the structural stability of fibre devices if applications are to be realized. For example, the electrochemically active materials coated on the fibre surface may peel off from the fibre substrate owing to the volume change after multiple charging and discharging cycles.

Energy harvesting devices

Flexible fibre-based solar cells can be made from both coaxial and twisted structures. To fabricate DSSCs with a coaxial structure, the semiconductor layer (typically TiO₂ nanoparticles or nanotubes), photoactive material (dye) and counter electrode shell (conducting polymer or carbon material) are sequentially deposited on a fibre electrode¹⁹ (FIG. 3a), followed by injection of a redox electrolyte. In twisted structures, the fibre photoanode that has been deposited with a semiconductor layer and coated with dye is wound with a fibre counter electrode^{20,21} (FIG. 3b). More specifically, dye-adsorbed TiO₂ nanoparticles are deposited on an aligned MWCNT fibre as the photoanode, which is then twisted with another aligned MWCNT fibre counter electrode using the above-mentioned rotation-translation setup (FIG. 1b). After incorporation of a redox liquid electrolyte, the resulting fibre-based DSSC shows a power conversion efficiency of 2.94%²¹.

Fibre-based solar cells in both coaxial and twisted structures demonstrate high flexibility. For example, the curves of current density as a function of voltage for the twisted DSSC are approximately equivalent before and after bending⁴², and the power conversion efficiency of a coaxial fibre-shaped DSSC is maintained by 95% after

bending for 100 cycles¹⁷. If the fibre electrode is elastic and designed as a spring, the DSSC can be stretched to form a close fit with the curved surface of the human body⁹. In addition, the incorporation of another functional material with carbon nanomaterial fibres can result in a flexible DSSC with more diverse functionalities. For example, MWCNT bundles are pulled through dispersions of either Fe₃O₄ or nickel nanoparticles to form hybrid fibres that are then used as counter electrodes in DSSCs³⁹ (FIGS 3c,d). The hybrid counter electrode is then wound onto a photoanode fibre (a dye-adsorbed TiO₂ nanotube grown on a titanium wire) using a rotation-translation setup to create a 1D DSSC (FIG. 3e). Under the influence of an external magnetic field, the DSSC can attach and detach from a substrate, which is promising for smart devices in, for example, the aerospace field.

Power conversion efficiency is a key factor in the development of photovoltaic devices, and it is particularly important for fibre-based solar cells. A high power conversion efficiency guarantees an acceptable output of electric power. Thus, research has been focused on making fibre electrodes with high mechanical, electrical and electrochemical properties for high power conversion efficiencies⁴³. It is generally realized by incorporating high-performance components to enhance the above-mentioned properties of the resultant fibre electrodes. For example, platinum nanoparticles are widely deposited to enhance the electrochemical activity of MWCNT fibre electrodes. This hybrid fibre electrode achieved a much higher power conversion efficiency of 8.10% in comparison to 4.91% derived from a bare MWCNT fibre⁴⁴.

There are currently three main types of fibre electrode: metal wires, modified chemical fibres and carbon nanostructured fibres. Metal wires, such as titanium⁴⁵, aluminium⁴⁶ and stainless steel wires²⁰, have the highest electrical conductivity, which is an advantage for the production of devices that are kilometres in length, but generally have poor electrochemical activity. Although it can be enhanced by incorporating more electrochemically

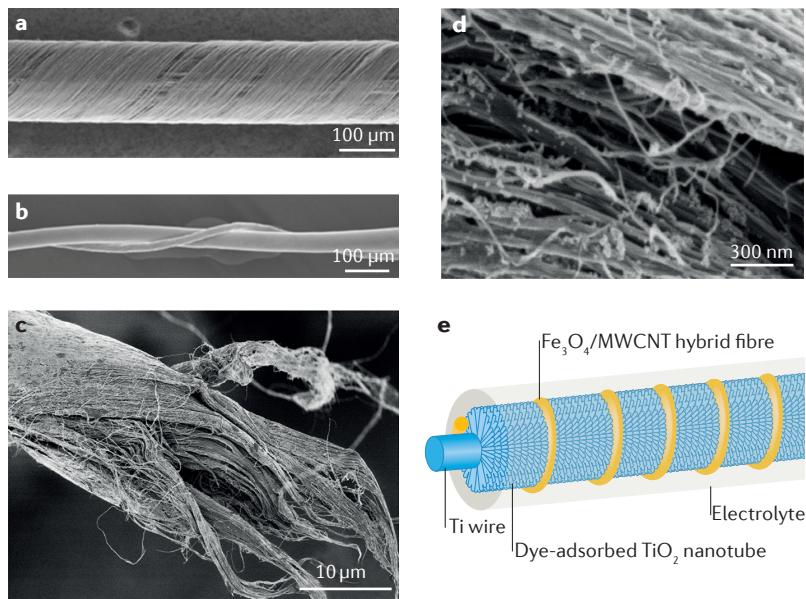


Figure 3 | 1D solar cells. **a,b** Scanning electron microscopy (SEM) images of coaxial and twisted dye-sensitized solar cells (DSSCs) in the absence of electrolytes. **c,d** SEM images of an Fe₃O₄/multi-walled carbon nanotube (MWCNT) hybrid fibre at low and high magnification, respectively. **e** A schematic illustration of the components of a Fe₃O₄/MWCNT DSSC fibre. Panel **a** is adapted with permission from REF. 19, Royal Society of Chemistry. Panel **b** is adapted with permission from REF. 21, American Chemical Society. Panels **c,d** and **e** are adapted with permission from REF. 39, Wiley-VCH.

active materials, such as platinum, conducting polymers and carbon nanomaterials, the smooth surfaces of the metal wires are unfavourable for high loadings of these additive materials. Consequently, a range of modification strategies exist to increase their surface roughness; for example, radially aligned mesopores and micropores are produced on aluminium wire via anodization and favour the deposition of graphene or MWCNTs⁴⁶. When the hybrid carbon–aluminium fibre is used as a counter electrode to fabricate a fibre-shaped DSSC, a power conversion efficiency of up to 6.8% is produced. Also, on a large scale, metal wires may be heavy and less suitable for portable or wearable products. To this end, natural or chemical fibres that may function as electrodes after coating a conducting layer on the surface are generally lightweight and flexible, which is favourable for large-scale production⁴⁷. However, the conducting layer may peel off from the chemical fibre under deformation⁴⁸. In solution-based processing routes, the smooth surfaces for chemical fibres are also not favourable for the deposition of active materials such as platinum, conducting polymers and carbon nanomaterials. Similar to metal wires, surface modifications by solvent or plasma treatment could be useful to enhance the surface roughness of the chemical fibre electrode for high loading capability.

Carbon nanomaterial fibres assembled from MWCNTs, graphene or their hybrids have shown high mechanical, electrical and electrochemical properties, and the nanostructured surfaces allow a more effective deposition of platinum⁴⁹, conducting polymers⁵⁰ or carbon nanomaterials⁵¹ to further enhance the electrical and electrochemical properties of the fibre electrode.

In addition, these carbon electrodes have large specific surface areas that promote the infiltration of active materials, resulting in electrodes with rapid charge transfer at the interface between electrode and active materials^{25,34,52}. For example, uniform and thin platinum nanoparticles are coated on a nanostructured graphene fibre. The graphene fibre is prepared from graphene oxide sheets in aqueous dispersion with the formation of a liquid crystalline phase, followed by reduction of graphene oxide⁵³. The graphene sheets are highly stacked and orientated along the fibre to make it mechanically strong and electrically conductive. Graphene sheets are ideal platforms for the uniform deposition of platinum nanoparticles with an average diameter of ~12 nm. When the platinum-deposited graphene fibre is used as a counter electrode, the resulting fibre-based DSSC demonstrates a certified power conversion efficiency of 8.45%⁴⁹. However, a lower power conversion efficiency of 5.97% is shown in the same DSSC when using a bulk carbon fibre electrode without a nanostructured surface. Much larger platinum crystals (up to hundreds of nanometres) are formed on the surface of this carbon fibre. These aggregated platinum crystals are also formed on metal wire (such as platinum wire) as the electrode, confirming the advantage of having a nanostructured surface for more effective deposition⁴⁹.

In general, photovoltaic devices must be sealed for practical application to avoid the influence of oxygen and moisture in air. In the case of fibre-based DSSCs, the photoanode and counter electrode can be inserted into a transparent heat-shrinkable tube that is then sealed at both ends after incorporation of the electrolyte. Other sealing strategies, such as spray coating of a sealing layer onto the device surface, may be promising in the future. In most cases, liquid electrolytes have been used to fabricate fibre-based DSSCs, resulting in complicated preparation procedures and reduced stability of the device during use. Hence, the use of gel-based or solid electrolytes is more attractive for wearable applications^{54,55}. It is currently challenging to find appropriate electrolytes to ensure that fibre-based DSSCs have high photovoltaic performance.

Sources of energy, other than solar energy, can be harvested on a fibre-based device. For example, fibre-based nanogenerators that use mechanical energy to produce electric energy have also been designed^{11,31,56–59}. In these nanogenerators, ZnO nanowires are used as the active piezoelectric material. ZnO nanowires are vertically grown on the surface of a Kevlar fibre, followed by coating with a metal such as gold. This coated fibre is then twisted together with an uncoated fibre³¹ (FIG. 4a). Charge is generated owing to a coupled piezoelectric and semiconducting property when the gold-coated electrode is stretched. For a brief time, a piezoelectric potential is generated on the gold-free ZnO nanowire under deformation. As a result, a Schottky barrier forms at the electrode interface, resulting in a flow of current from gold to ZnO (FIG. 4b). The output currents and voltages were observed at 4 nA and 1 mV, respectively. To further enhance the energy harvesting capability, some efforts have been made to design piezoelectric nanogenerators with a coaxial structure. Typically, ZnO nanowires (piezoelectric active layer) and

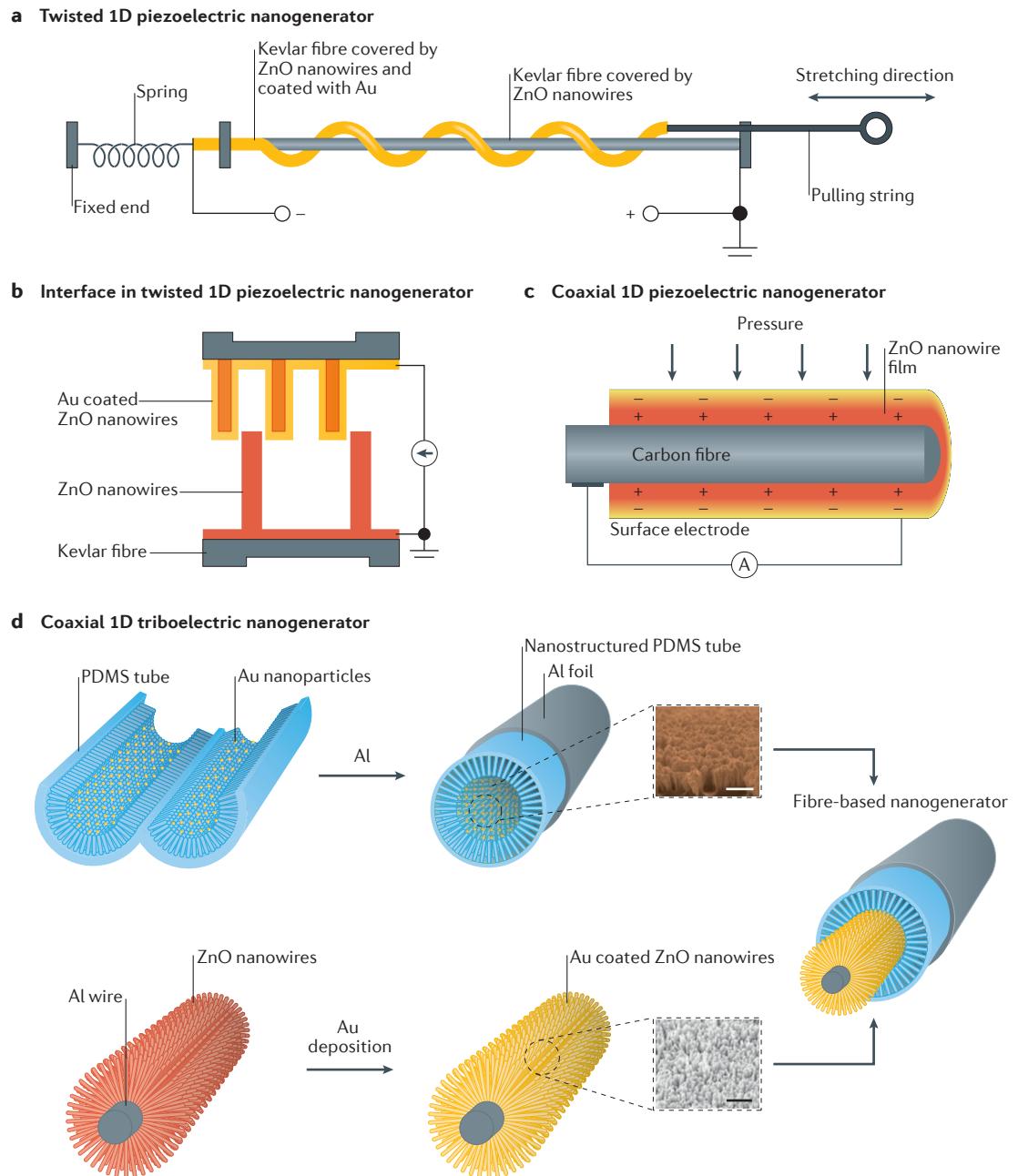


Figure 4 | Fibre-based piezoelectric and triboelectric nanogenerators. **a** | The experimental setup of a fibre-based piezoelectric nanogenerator with a twisted structure. **b** | The contact between the two fibres covered by ZnO nanowires. **c** | A coaxial fibre-based piezoelectric nanogenerator. Piezoelectric potential is generated by the ZnO nanowire film when pressure is applied. **d** | A coaxial fibre-based triboelectric nanogenerator. Electrons are generated at the interface between gold-coated ZnO nanowires and nanostructured polydimethylsiloxane (PDMS) under pressing. Panels **a** and **b** are adapted with permission from REF. 31, Macmillan Publishers Limited. Panel **c** is adapted with permission from REF. 56, Wiley-VCH. Panel **d** is adapted with permission from REF. 11, American Chemical Society.

a metal layer (surface electrode) are sequentially deposited on a carbon fibre (inner electrode) (FIG. 4c). Through the application of silver paste or the deposition of a gold layer, fibre-based piezoelectric nanogenerators based on ~1,000 carbon fibres are connected in parallel, which contributes to an output current and voltage of ~200 nA and ~3 V, respectively⁵⁶. The enhancement comes from both the increased number of devices and a more efficient

polarization of ZnO nanowires that occurs when pressure is applied; an enhancement of 7% for the output current is shown when the applied pressure increases by 35%.

The triboelectric effect — a type of contact electrification that uses the frictional contact of two different materials to generate electrons — is also used for energy harvesting in 1D systems. For example, a coaxial triboelectric nanogenerator is developed for enhancement of

the output current and voltage. Typically, polydimethylsiloxane (PDMS) nanowire arrays with a length of 1–2 µm are prepared inside an aluminium foil-covered PDMS tube as the outer electrode. A gold layer deposited on an aluminium wire that is coated with ZnO nanowires forms the inner electrode, which is inserted into the outer electrode to form a coaxial triboelectric nanogenerator (FIG. 4d). A rough interface is created between the gold-coated ZnO nanowires and the nanostructured PDMS, which enhances the contact of these materials at the triboelectric interface. As a result, the output current and voltage are enhanced to 10 µA and 40 V, respectively, which are much higher values compared with those derived from 1D piezoelectric nanogenerators^{31,56}.

Generally, both piezoelectric and triboelectric nanogenerators produce alternating currents and require rectification for many applications such as light-emitting diodes, electronic watches and mobile phones. In particular, the output current is still far from satisfactory. Hence, it is important for fibre-based nanogenerators to be integrated with energy storage devices: for example, supercapacitors, to enhance and stabilize the output current. To this end, 2D systems may offer some prototypes for structure design; a planar triboelectric nanogenerator and supercapacitor can be sequentially built on a stretchable rubber substrate and connected with a rectifier for production of a self-powered energy module⁶⁰. Based on a similar design, producing wearable power textiles is accessible by stacking two textiles woven with a fibre-based triboelectric nanogenerator and supercapacitor. In addition, these two kinds of fibres could be woven into one textile as the warp and weft yarns, which can reduce the thickness and enhance breathability of the textiles compared with the method of stacking two textiles. Furthermore, the high level of integration obtained from incorporating both fibres in the same textile may contribute to structural stability under deformation, and hence represents a promising direction in the future.

Energy storage devices

At present, the existing range of 1D energy storage devices includes supercapacitors^{22–24,28,46,61–70}, lithium-ion batteries^{34,71–75}, lithium–sulfur batteries³⁶, lithium–air batteries³⁷, zinc–air batteries³⁵ and aluminium–air batteries³⁸. Of these, supercapacitors and lithium-ion batteries are outlined in this Review.

Coaxial and twisted structures are both also applicable for fibre-based supercapacitors. A large specific surface area is particularly important for the fibre electrode to achieve high electrical double layer capacitances owing to more sites for ion adsorption at the electrode/electrolyte interface. Therefore, the materials that are of principal interest are various carbon nanomaterial fibres based on MWCNT^{66,76–79} and graphene^{68,80,81}, as well as their composites with conducting polymers or metal oxides. In a typical twisted structure, polyaniline is electrochemically deposited on aligned MWCNT fibres, which are then coated by a layer of gel electrolyte. Two of these gel-coated hybrid fibres composed of polyaniline and MWCNT are wound together to form a fibre-based supercapacitor⁵⁰ (FIG. 5a). For a coaxial structure, an aligned MWCNT fibre and

sheet serve as the inner and external electrodes with a layer of gel electrolyte sandwiched between them^{15,53} (FIG. 5b). The twisting structure is more favourable for rapid and continuous fabrication for large-scale production of fibre-based supercapacitors⁵¹, whereas the coaxial structure provides higher stability under deformation owing to better contact between the electrode and electrolyte¹⁵ (FIG. 5c).

In lithium-ion batteries, it is technically challenging to efficiently incorporate electrochemically active materials, such as $\text{Li}_4\text{Ti}_5\text{O}_{12}$ and LiMn_2O_4 , into the external electrode in a coaxial structure. As a result, fibre-based lithium-ion batteries are currently made by depositing the active materials into two fibre electrodes, followed by twisting the fibres together. In contrast to the incorporation methods used for conventional lithium-ion batteries, the active materials are scrolled into fibre electrodes⁸². In this system, aligned MWCNT sheets that are porous and strong are first paved on a smooth substrate, followed by deposition of active materials. The resulting hybrid sheets are then scrolled to form a hybrid fibre⁸².

Based on a similar strategy, $\text{Li}_4\text{Ti}_5\text{O}_{12}$ and LiMn_2O_4 nanoparticles are scrolled into MWCNTs at high weight percentages of 78% and 90%, respectively⁷¹ (FIG. 5d). The two hybrid fibres serve as the anode and cathode of a lithium-ion battery, and LiPF_6 in a solvent mixture of ethylene carbonate, diethyl carbonate and dimethyl carbonate is used as the electrolyte of the battery. High energy and power densities of 17.7 mWh cm^{-3} and 0.56 W cm^{-3} have been produced⁷¹. However, it remains difficult to scale up the preparation of the hybrid fibre electrode. More efforts should be made to develop new conducting materials that are generally 3D interpenetrating carbon networks to host active materials, and to design new preparation methods, such as wet spinning, to produce continuous hybrid fibres with high capacities.

Fibre-based supercapacitors and lithium-ion batteries are flexible and can be bent, twisted and tied into various shapes without damaging the materials or showing any decreases in properties. For example, the specific capacitance of a fibre supercapacitor is maintained at 96.6% after bending for 100,000 cycles⁸. By using an elastomeric fibre or designing a spring structure, fibre-based energy storage devices can be made to stretch up to 400%^{83,84}. After incorporation of a second functional phase into the electrode, the resulting energy storage devices demonstrate more functionalities besides storing energy, for example, self-healing, colour change in response to external stimuli and shape memory^{26–28,69,85–87}. A self-healing fibre electrode can be prepared by wrapping the aligned MWCNT sheet onto a self-healing polymer fibre and then twisting these two composite fibres together to form a fibre supercapacitor²⁶ (FIG. 5e). This supercapacitor self-heals after breaking if the fibres are reconnected at the breaking site. At the cross-section of the breaking site, the formation of hydrogen bonds recovers the mechanical properties of the fibre, and the van der Waals interactions between neighbouring MWCNTs lead to the reconnection of the conducting network. In another example, chromatic polymers, such as polyaniline, that display different colours under charging or discharging have been

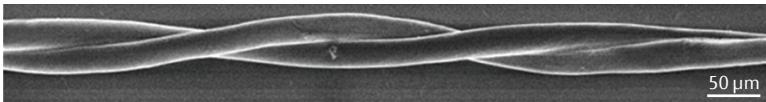
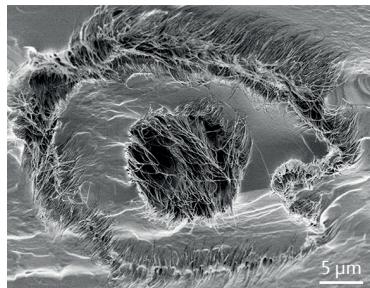
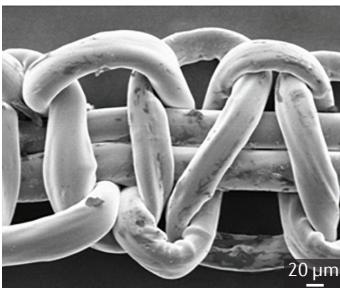
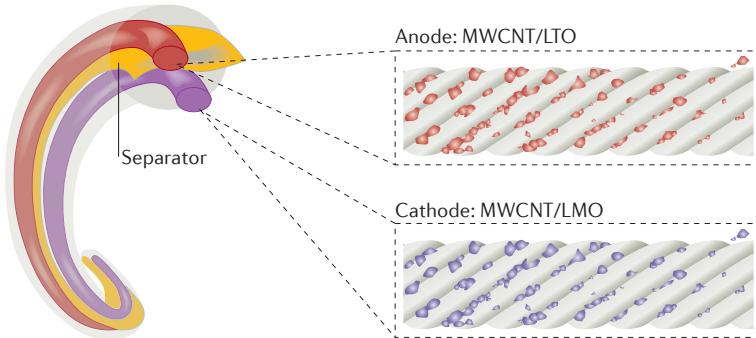
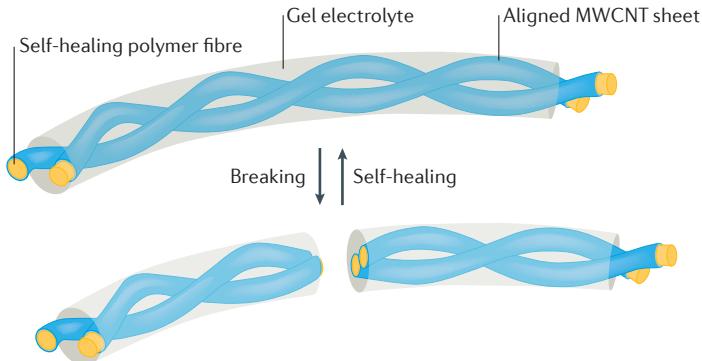
a Twisted 1D supercapacitor**b Coaxial 1D supercapacitor****c Knitted coaxial 1D supercapacitor****d 1D lithium-ion battery****e Self-healing 1D supercapacitor**

Figure 5 | 1D supercapacitors and lithium-ion batteries. **a,b** Scanning electron microscopy (SEM) images of twisted and coaxial supercapacitors. **c** SEM image of a coaxial supercapacitor in a knitted conformation. **d** A schematic illustration of a 1D lithium-ion battery. Multi-walled carbon nanotube/Li₄Ti₅O₁₂ (MWCNT/LTO) and MWCNT/LiMn₂O₄ (MWCNT/LMO) function as the anode and cathode, respectively. **e** A schematic illustration of a self-healing supercapacitor. Aligned MWCNT sheets are wrapped onto the surface of the self-healing polymer fibre as the electrode. Panel **a** is adapted with permission from REF. 50, Royal Society of Chemistry. Panels **b** and **c** are adapted with permission from REF. 15, Wiley-VCH. Panel **d** is adapted with permission from REF. 71, Wiley-VCH. Panel **e** is adapted with permission from REF. 26, Wiley-VCH.

incorporated into aligned MWCNT fibre electrodes in a fibre supercapacitor²⁷. This colourful display enables the working state of the fibre supercapacitor to be easily judged by directly observing the chromatic transition.

Integration of 1D structures

The 1D configuration offers some unique and attractive features in energy harvesting and storage. The integration of 1D energy harvesting and storage components in one device that can convert environmental energy into electrical energy and simultaneously store energy allows more convenient and efficiently managed energy devices. Once they are weaved together to make fabrics, such devices can be connected in series or parallel to enhance the output voltage or current, respectively. Therefore, although 1D energy harvesting or storage devices typically output a limited voltage or current, an integrated system can be designed to satisfy a range of applications. For example, the working voltages of fibre-based supercapacitors are generally hundreds of millivolts. Inspired by the assembly of electrocytes in electric eels, the connection, in series, of many fibre-based supercapacitors along an aligned MWCNT composite fibre results in voltages as high as 1,000 V (REF. 8) (FIG. 6a). This integrated supercapacitor design also provides high flexibility and stability on bending and stretching. Impressively, the specific capacitances are maintained after deforming for 100,000 cycles.

A supercapacitor displays high power density and low energy density, but a lithium-ion battery displays low power density and high energy density. It remains challenging to simultaneously achieve high power and energy densities in a single energy storage device. Three fibre electrodes could be twisted together to produce a 1D energy storage device that can function as both a supercapacitor and lithium-ion battery by sharing a hybrid fibre as the common electrode⁸⁸ (FIG. 6b). In other words, when the switch that connects the aligned MWCNT/ordered mesoporous carbon and aligned MWCNT/Li₄Ti₅O₁₂ is on, a high power density is generated; when the switch that connects the aligned MWCNT/LiMn₂O₄ and aligned MWCNT/Li₄Ti₅O₁₂ is on, a high energy density is generated. The power and energy densities attained are 1 W cm⁻³ and 50 mWh cm⁻³, respectively. In addition, the supercapacitor component can be self-charged by the lithium-ion battery. Based on a similar strategy, more hybrid fibres with additional functionalities could be twisted into a 1D configuration, which represents a promising direction for the future.

The integration of energy harvesting and storage devices is generally realized by coating in turn photovoltaic and electrochemically active materials on a fibre electrode or sequentially depositing them from inside out^{29,30}. For example, a semiconductor layer containing a photoactive dye and an electrochemically active material were coated on the left and right parts of a titania-modified titanium wire that served as the common electrode. Two aligned MWCNT fibre electrodes were then wound around both parts and coated with appropriate electrolytes, producing energy harvesting and storage parts at the left and right of the fibre, respectively²⁹ (FIG. 6c). Under illumination, the DSSC component can charge the supercapacitor to 0.6 V within seconds. In recent years, for the further enhancement of the energy storage capability of the supercapacitor component, various pseudocapacitance materials have been incorporated

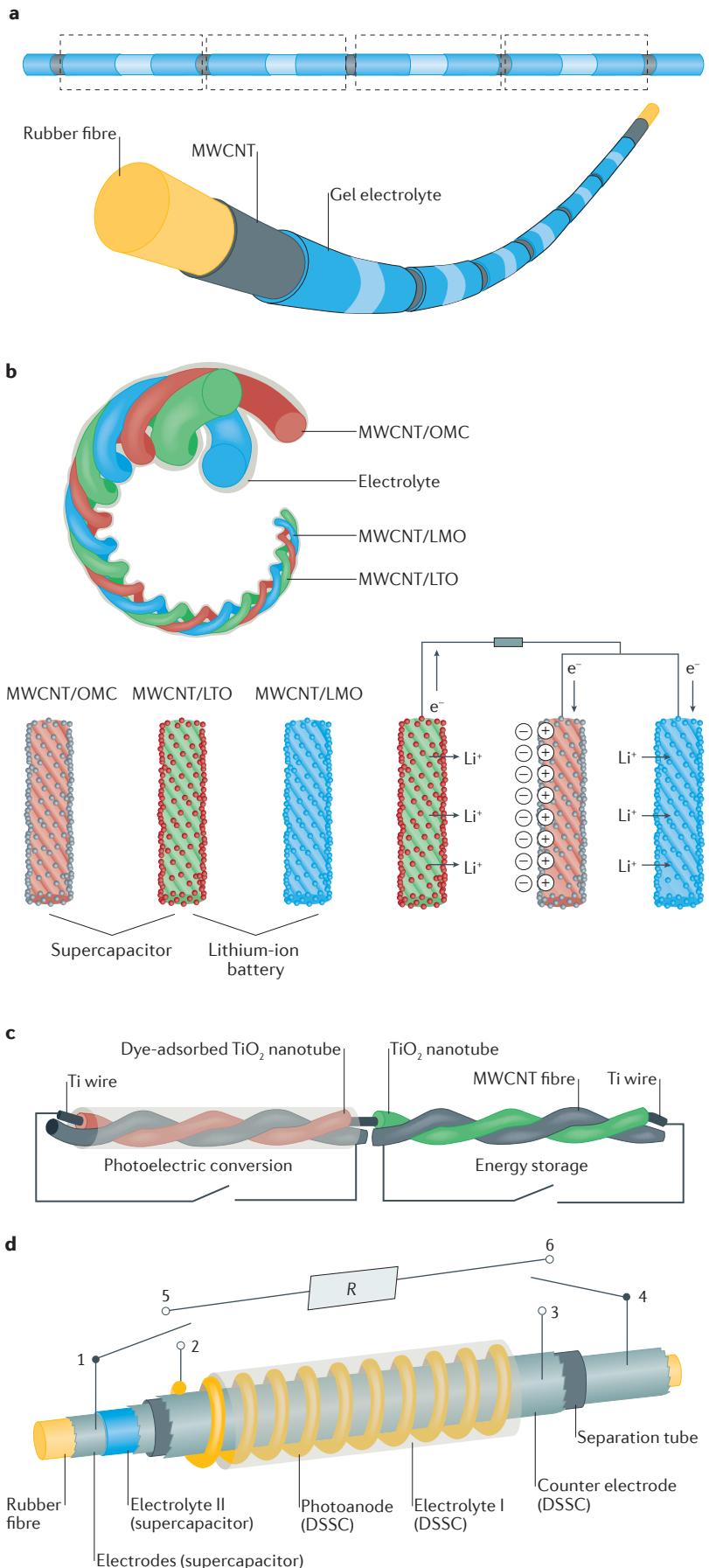


Figure 6 | Assembly and integration of 1D energy harvesting and storage devices. **a** | A 1D supercapacitor based on in-series connections. The energy storage units share the inner electrode. **b** | A hybrid energy storage device formed by twisting three fibre electrodes together. The multi-walled carbon nanotube/Li₄Ti₅O₁₂ (MWCNT/LTO) electrode serves as a common electrode for a lithium-ion battery and supercapacitor. During the discharge process, the MWCNT/LiMn₂O₄ (MWCNT/LMO) and MWCNT/ordered mesoporous carbon (MWCNT/OMC) electrodes are connected to function as positive electrodes, and the MWCNT/LTO electrode serves as the common negative electrode, by which the hybrid device can discharge at high current densities. **c** | Integrated energy harvesting and storage devices with an in-series architecture. Photoelectric conversion and energy storage components are produced on the left and right of a fibre, respectively. Under light illumination, the energy storage component is photocharged by the photoelectric conversion component with the left and right switches kept at 'on' and 'off' states, respectively; the discharging process begins when the left and right switches are changed to 'off' and 'on' states, respectively. **d** | Integrated energy harvesting and storage device with a core–shell architecture. Energy storage and harvesting components are sequentially deposited on the surface of a fibre. The energy storage component is photocharged by the energy harvesting component under light illumination when electrodes 1 and 4 are connected with electrodes 2 and 3, respectively; the discharging process performs when electrodes 1 and 4 are connected with electrodes 5 and 6, respectively. *R*, resistance in external circuit. Panel **a** is adapted with permission from REF. 8, Wiley-VCH. Panel **b** is adapted with permission from REF. 88, Wiley-VCH. Panel **c** is adapted with permission from REF. 29, Wiley-VCH. Panel **d** is adapted with permission from REF. 30, Wiley-VCH.

into the aligned MWCNT sheet^{89,90}. In another study, a lithium-ion battery can be integrated with a photovoltaic component for a much higher energy density owing to the similar in-series organization⁹¹. This results in an increase in photocharging duration by two to three orders of magnitude. In addition to the typical in-series architecture, it is also possible to make an integrated device with a core–shell structure by fabricating the energy storage core on a fibre substrate and then preparing the energy harvesting shell³⁰ (FIG. 6d). The electric energy generated by the DSSC shell can be simultaneously stored by the supercapacitor core in electrochemical energy form.

Interfaces within 1D electrodes and devices

Alongside optimization of the material and the assembled structure in the power system, it is important to understand the interfaces within the configuration to enhance the energy harvesting and storage capability. The interfaces between active material and electrolyte, between active material and electrode, and between the neighbouring building blocks of the fibre electrode are outlined in this section. For example, the interface between photoactive material and curved fibre electrode proves critical for charge transfer, which influences the power conversion efficiencies of 1D organic solar cells.

In addition, the interface between aligned building blocks, such as CNTs, of the fibre electrode is particularly important for charge transport in 1D systems, owing to the much longer charge transport pathway along the fibre electrode compared with planar counterparts. These interfaces — on the curved fibre substrates — are different from their widely studied 2D and 3D counterparts.

For example, it has been theoretically predicted that the photovoltaic performance of perpendicularly aligned TiO_2 nanotubes will exceed the performance of TiO_2 nanoparticles at the photoanode of a DSSC. This is because charges have to diffuse across many boundaries that are formed by nanoparticles, whereas charge transport in nanotubes is rapid. However, in conventionally planar structures, the use of aligned TiO_2 nanotubes generally results in lower power conversion efficiencies compared with nanoparticles^{92,93}. This differing observation to the theoretical prediction may be attributed to the densely packed arrangement of the perpendicularly aligned TiO_2 nanotubes, which results in limited infiltration of the electrolyte to the top part of the arrangement, leaving air trapped beneath. In other words, the specific surface area of the aligned TiO_2 nanotubes is only partially used for charge transfer. By contrast, TiO_2 nanotubes grown on the surface of titanium wire form V-shaped voids, and the electrolyte, as well as dye molecules, can effectively infiltrate from the broad top voids of the TiO_2 nanotubes to the narrow bottom^{45,49} (FIG. 7a). In contrast to the planar structures, the fibre-based DSSCs with aligned TiO_2 nanotubes demonstrate a much higher power conversion efficiency compared with TiO_2 nanoparticles^{21,49,94,95}. Therefore, by optimizing the interface between active material and electrolyte through the design of TiO_2 nanotubes with balanced density and diameter, electrolyte infiltration into the dye-adsorbed TiO_2 nanotubes can be further enhanced, thus promoting charge transfer for higher power conversion efficiency.

However, the curved surfaces of fibres have proven tricky substrates for the deposition of active materials. For example, high-quality thin perovskite layers are needed to achieve high power conversion efficiencies in solar cells, and such layers have been difficult to obtain as conventional planar substrates have been replaced with fibre electrodes. Low-quality perovskite crystals are observed to poorly coat a metal wire using a typical dip-coating method because of the smooth surface of the wire, and hence power conversion efficiencies are below 3.3%¹⁸. A range of deposition methods developed since have resulted in the enhancement of layer continuity and uniformity^{96,97}. A cathodic deposition method has been shown to prepare uniform perovskite layers. More specifically, a porous PbO layer deposited on a curved titanium wire coated with TiO_2 nanotubes reacts with hydroiodic acid and $\text{CH}_3\text{NH}_3\text{I}$ in sequence to form nanocrystals of the perovskite $\text{CH}_3\text{NH}_3\text{PbI}_3$ with high coverage and uniformity. This method enhances the power conversion efficiency to 7.1%⁹⁷. Considering flexible 2D perovskite solar cells have achieved power conversion efficiencies of 16%⁹⁸, there is scope to further increase the photovoltaic performance of fibre-based perovskite solar cells.

Charge transport is critical for all energy harvesting and storage devices⁹⁹, but it is particularly important for 1D devices, because the transport pathway in the fibre electrode can be several orders of magnitude longer than that in conventional planar electrodes. The fibre electrodes based on carbon nanomaterials — including MWCNT and graphene — have been mostly explored to understand and optimize the charge transport in energy harvesting and storage.

If MWCNTs and graphene are building blocks for fibre electrodes, their size and orientation are key factors in determining the performances of these electrodes. For MWCNTs and graphene fibres, the electrical resistances are mainly derived from their contact resistances — this can be inferred because resistances for individual MWCNTs and graphene sheets are very low. The size and degree of orientation can considerably affect the contact resistance. For example, if longer MWCNTs and larger graphene sheets are used to build the same length of fibre electrode, there will be fewer interfaces among them and the contact resistance will decrease. More specifically, the electrical conductivities of graphene fibres were enhanced from 250 to 390 S cm^{-1} by increasing the length of the graphene sheet from 0.84 to 18.5 μm (REF. 100). For a MWCNT fibre, the charges can rapidly hop from the outer MWCNTs to the inner ones based on a 3D hopping conduction model and can then effectively transport along the axial direction of the fibre^{101,102} (FIG. 7b). Decreasing the distances between MWCNTs can promote hopping efficiency and enhance charge transport. The pressing and annealing treatment has been conducted to enhance compactness to produce electrical conductivities of up to $2.24 \times 10^4 \text{ S cm}^{-1}$ (REFS 103,104). However, it remains challenging to wet-spin larger graphene sheets and dry-spin longer MWCNTs in the preparation of the fibre electrode¹⁰⁵.

The alignment of the MWCNTs or graphene sheets is important for rapid charge transport^{16,22,52,106–113}. Compared with MWCNT networks that contain many boundaries, the alignment of MWCNTs in the fibre electrode can largely enhance the charge transport according to the 3D hopping conduction model. The charges can more effectively transport along the aligned MWCNTs without the delays produced by the boundaries in MWCNT networks¹¹⁴. The alignment of the carbon nanomaterials also induces the orientation of the incorporated active materials^{50,115}, and the charge transport between the active material and carbon nanomaterial become more effective. For example, polyaniline was grown along aligned MWCNTs, and the composite fibre electrodes contributed greatly enhanced specific capacities in the resulting fibre supercapacitor⁵⁰.

Conclusions and outlook

In the past decade, the interdisciplinary research area of 1D energy harvesting and storage devices has rapidly developed to become a promising branch of modern electronics. However, challenges remain that hinder the practical applications of 1D devices — in particular, related to their poor performance. For example, perovskite solar cells using 1D configurations show power

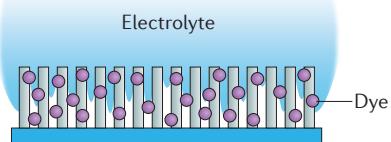
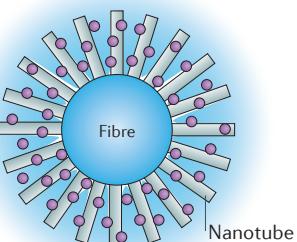
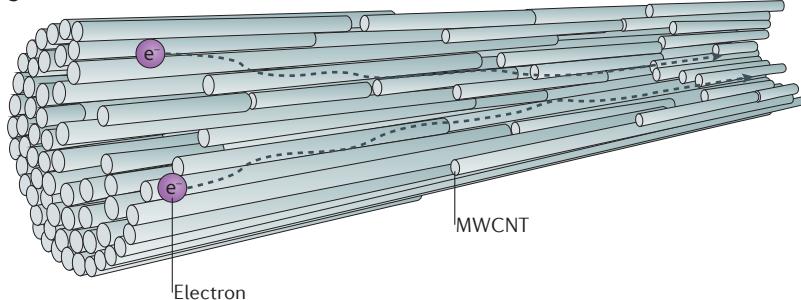
a 2D configuration**1D configuration****b**

Figure 7 | Interfaces within 1D DSSCs and aligned MWCNT fibres. **a** | Electrolyte infiltration into dye-sensitized TiO_2 nanotubes differs in 2D and 1D configurations. In 2D structures, electrolyte infiltration is hindered by the dense TiO_2 nanotubes. However, in 1D arrangements, the curved fibre substrate contributes to the formation of V-shaped voids, which favour electrolyte infiltration. **b** | A model of electron hopping within a conducting fibre made from aligned multi-walled carbon nanotubes (MWCNTs). The dashed line indicates the transport pathway of electrons. The ability of electrons to hop at the interfaces between neighbouring MWCNTs is crucial in determining the electrical resistance of the fibre electrode. Fewer interfaces and shorter distances at the interfaces help to reduce the electron loss during the hopping process. DSSCs, dye-sensitized solar cells.

conversion efficiencies of 7.1%, which is considerably lower than efficiencies of around 16% for their flexible planar counterparts^{97,98}. These lower photovoltaic efficiencies are a result of the difficulty of forming thin and uniform perovskite layers using a metal as a fibre electrode. Perovskite solar cells fabricated using fibres of carbon nanomaterials show electrical conductivities ranging from 10^2 to 10^3 S cm^{-1} , which are far too low for continuous production. Hence, electrical conductivities need to be enhanced by two orders of magnitude, while maintaining the voided structure, good mechanical properties and high flexibility. Although several examples of interface optimization have been demonstrated, they are applicable to a limited number of material systems, and there is a lack of systematic and in-depth understanding of the mechanisms involved.

More research should focus on exploring new electrode and active materials to enhance the performances of 1D energy harvesting and storage devices¹¹⁶. For example, single-walled CNTs are promising materials to enhance charge transport for highly conductive fibre electrodes; 2D materials, such as graphene oxide and MoS_2 , can be explored as active materials to enhance the energy storage performance of the obtained devices. A systematic study of the mechanisms and underlying relationships, such as the dependence of loading density

and alignment degree on the curvature derived from the fibre configuration, is desired to improve the energy harvesting or storage performance by optimizing the interface and structure.

The structural stabilities of the component materials and the resultant assemblies are crucial factors that determine the lifetime of 1D energy harvesting and storage devices. The fibre electrodes need to be sufficiently tough to avoid breaking under deformation, and the interfaces between electrodes and the active material need to be stable enough to prevent the active material from peeling off of the electrode. Hence, the ability to stretch these devices has been extensively studied to ensure the devices can accommodate the accompanying deformation under shearing during use^{9,23,30,35,74,77,78,83,117–119}. However, the long-term stability of these 1D energy harvesting and storage devices is much lower than expected, despite the realization of various stretchable systems. The use of liquid electrolytes further reduces the stability in many 1D energy harvesting and storage devices, because effective long-term encapsulation of liquids in a fibre system is still technically difficult. Mainstream encapsulation in plastic tubes also decreases the flexibility and performance of the resulting fibre device. On one hand, it is extremely important to find new and efficient encapsulation methods for a continuous fabrication that applies to current industrial production. On the other hand, stable and environmentally friendly gel electrolytes that are suitable for fibre substrates should be extensively investigated to advance their practical applications. Recent progress has been made on the synthesis of highly stable gel electrolytes. For example, a polymer–ionic liquid gel with high non-volatility and durability has been introduced to 1D DSSCs⁵⁴. Poly(vinylidene fluoride-co-hexafluoropropene) (PVDF-HFP) copolymer serves as the gelling agent, and 1-butyl-3-methylimidazolium bis(trifluoromethanesulfonyl) imide (BMITFSI) provides both high ionic conductivity and hydrophobic capability to the electrolyte. Hence, the resulting 1D DSSCs demonstrate an impressively high stability with 90% of the initial power conversion efficiency remaining after 30 days in comparison to only 20% when using a conventional liquid electrolyte⁵⁴.

To fully realize the commercial potential of 1D energy harvesting and storage devices, the development of efficient weaving techniques is essential for the production of flexible, breathable and wearable textiles. However, 1D devices that are currently available have been mainly woven into textiles by hand^{8,11,89,120–122}. To promote their large-scale application, machine-based weaving techniques must be developed^{10,12}. On the basis of the well-developed weaving technologies, some small-sized power textiles can be produced. For example, a loom is used to weave seven in-series 1D supercapacitors into a $15 \times 10 \text{ cm}$ power textile with a capacitance of 14 mF (REF. 10). Within a textile or fabric to be used for energy harvesting or storage applications, the optimal way of connecting many fibre electrodes must be devised, and to the best of our knowledge only a few studies are available. Moreover, the multifunctionalization and integration of 1D devices is vital for reaching a broader range of applications. For example, 1D supercapacitors

that can self-heal may avoid failure due to breaking and thus demonstrate a better adaptability in harsh environment^{26,123}. In addition, integration of 1D devices, such as supercapacitors and sensors, can enable the ability to monitor health conditions of the human body without use of an external power source¹²⁴. For wearable electronics, there are, of course, other considerations, such as comfort and washability, that should be investigated as the range of textiles becomes more diverse and their performance improves.

The main concern for 1D devices is how to reach high efficiencies in the near future. We can expect that future clothes will be made from these energy harvesting and storage fibres and will be used as dominating power sources in our daily life, which may bring a technical revolution to a certain degree.

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