Direct coherent multi-ink printing of fabric supercapacitors

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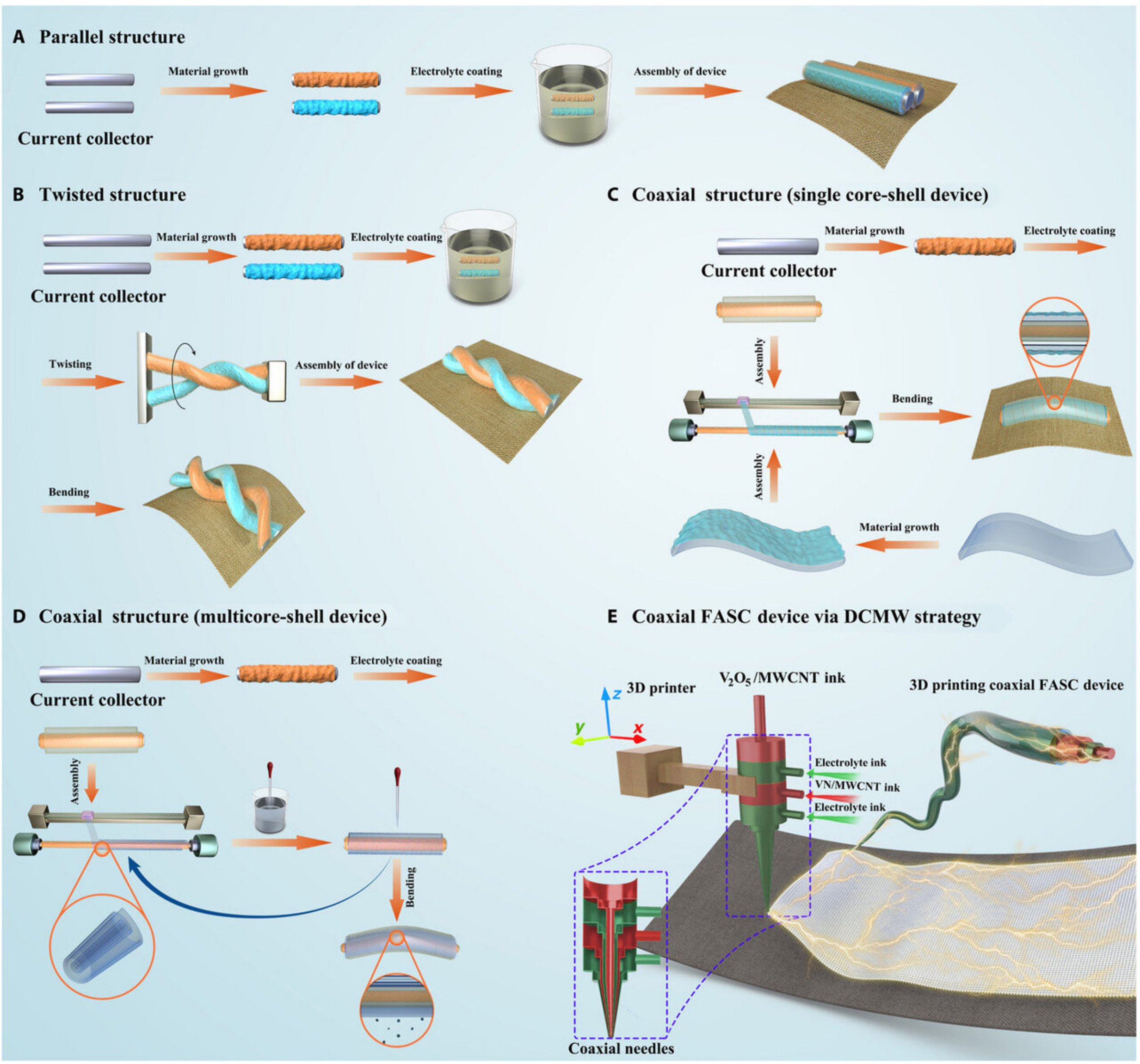


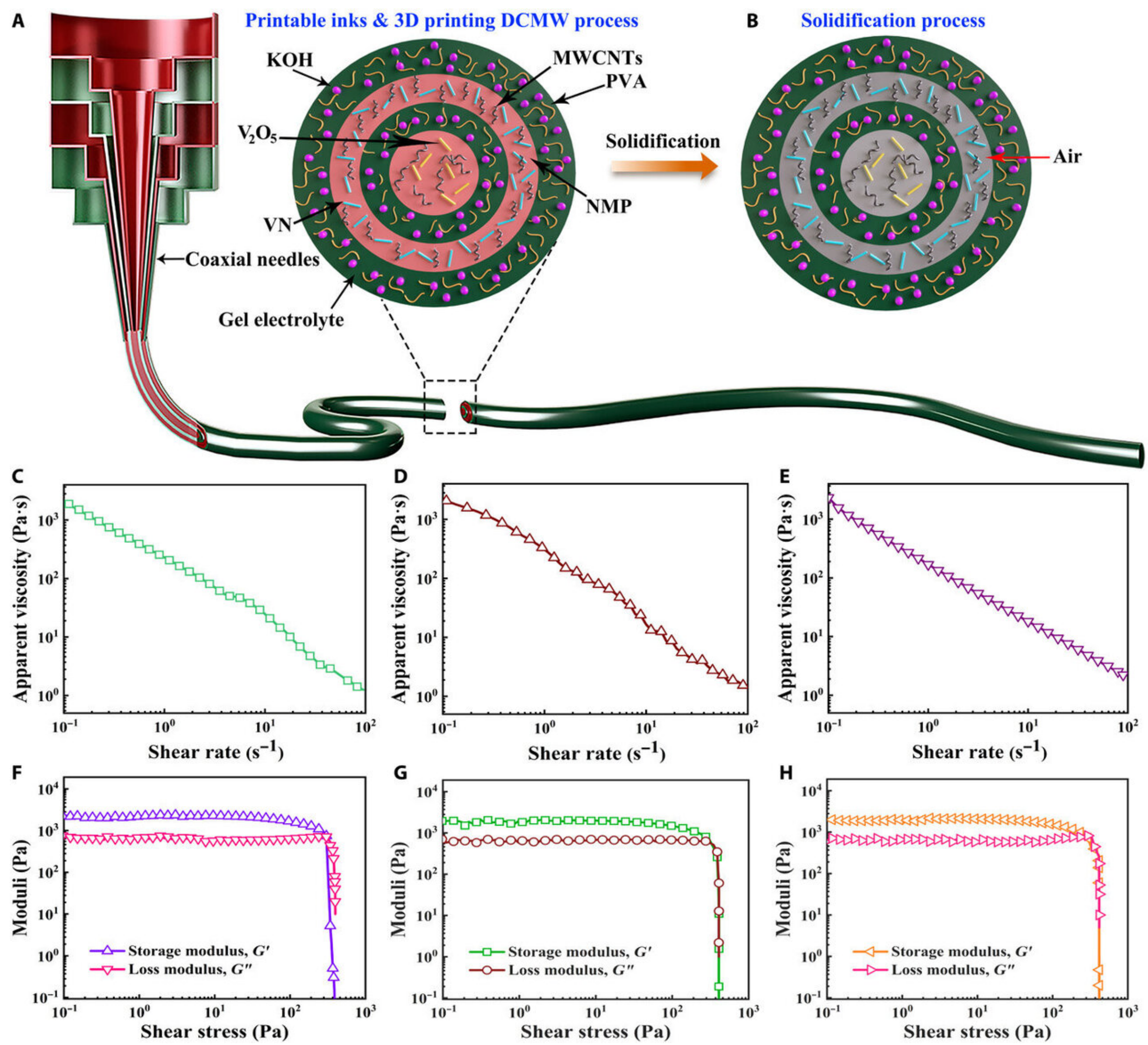
Diagrammatic illustration of the fabrication process of various FASC devices.

Schematic diagram of the comparison of the preparation process of the conventional FASC device with (A) parallel, (B) twisted, (C and D) coaxial architectures, and (E) our development of three-dimensional (3D) printing coaxial FASC device via a direct coherent multi-ink writing (DCMW) technology. Credit: Science Advances, doi: 10.1126/sciadv.abd6978

Fiber-shaped supercapacitors are a desirable high-performance energy storage technology for wearable electronics. The traditional method for device fabrication is based on a multistep approach to construct energy devices, which can present challenges during fabrication, scalability and durability. To overcome these restrictions, Jingxin Zhao and a team of scientists in physics, electrochemical energy, nanoscience, materials, and chemical engineering in China, the U.S., and Singapore, developed an all-in-one coaxial fiber-shaped asymmetric supercapacitor (FASC) device. The team used direct coherent multi-ink writing, three-dimensional (3-D) printing technology by designing the internal structure of the coaxial needles and regulating the rheological property and feed rates of the multi-ink. The device delivered a superior areal energy and power density with outstanding mechanical stability. The team integrated the fiber-shaped asymmetric supercapacitor (FASC) with mechanical units and pressure sensors to realize high performance and self-powered mechanical devices to monitor systems. The work is now published on *Science Advances.*

Texture-based wearable electronics

Advances in textile-based wearable electronics can be achieved with advanced fibrous energy storage devices with excellent knittability, flexibility and high mechanical stability. Fiber-shaped asymmetric supercapacitors (FASCs) are widely in use to develop wearable electronics as a promising fiber-shaped energy storage device due to their high power density, long cycling stability, excellent reversibility and improved energy density. In this work, Zhou et al. integrated high-throughput 3-D printing direct ink-writing technology to construct the all-in-one coaxial FASC device with compact internal structures. For this, they rationally designed the device using 3-D printed direct, coherent multi-ink writing (DCMW). The team also designed the internal structure of the multicore-shell needles by charge matching different electrodes, where the rheological properties of the multi-inks matched each other from the innermost layer to the outermost layer during 3-D printing.

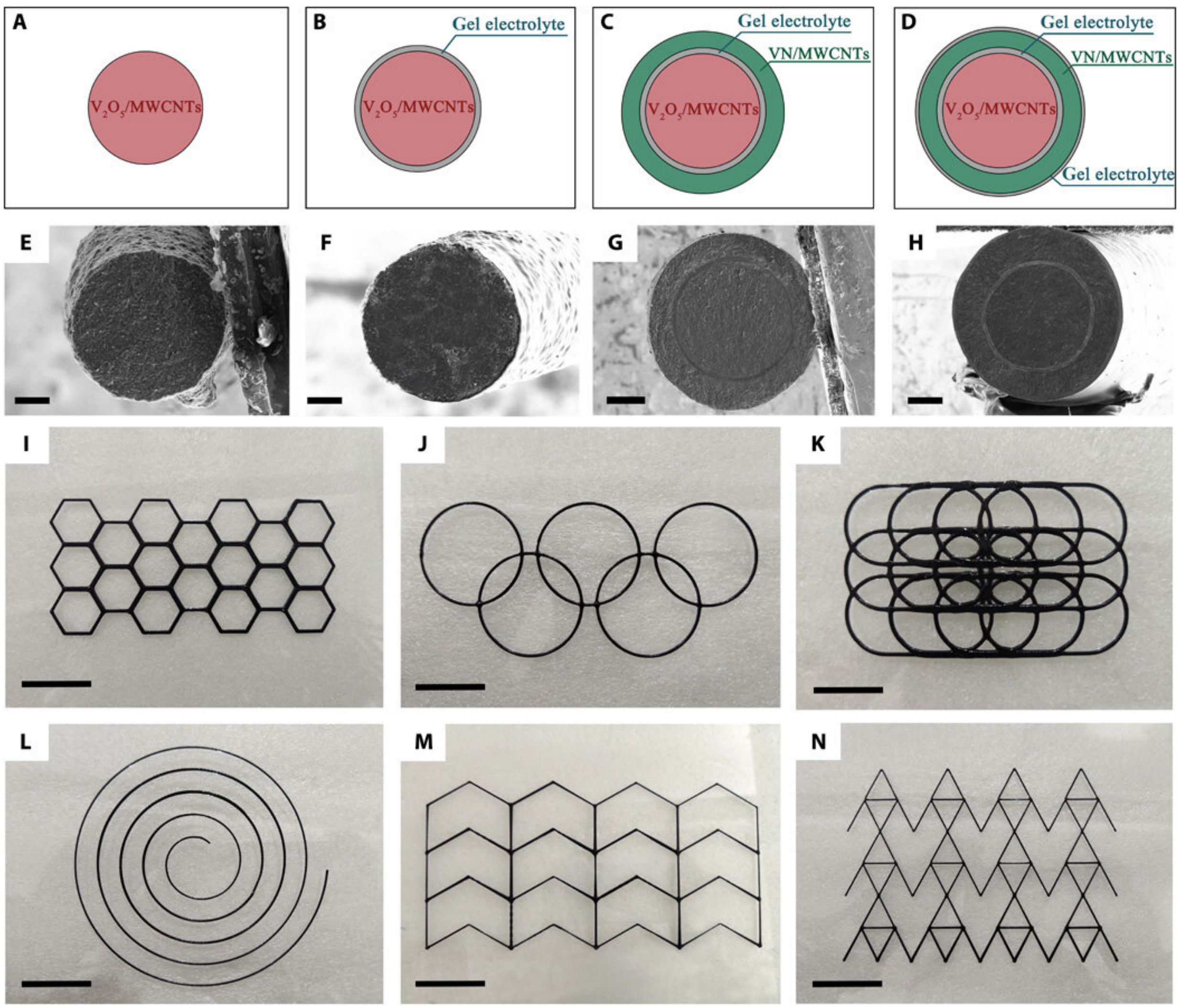


Rheological performance of the as-fabricated inks. (A) 3D printing extrusion process of the printable coaxial FASC device. (B) 3D printing coaxial FASC device is achieved by subsequent solidification process. Rheological properties of pure MWCNT, V2O5 NW/MWCNT, and VN NW/MWCNT slurry inks. (C to E) Apparent viscosity as a function of shear rate for pure MWCNT, V2O5 NW/MWCNT, and VN NWs/MWCNT inks, respectively. (F to H) Storage modulus, G′, and loss modulus, G″, as a function of shear stress for pure MWCNT, V2O5 NW/MWCNT, and VN NW/MWCNT slurry inks, respectively. Credit: Science Advances, doi: 10.1126/sciadv.abd6978

The device contained a compact four-layer structure that shortened the ion diffusion path to improve the electrochemical performance and mechanical durability of the device under bending. The team produced a proof-of-concept FASC device with vanadium oxide nanowires/ multiwalled carbon nanotubes (MWCNTs) and vanadium nitride (VN) nanowires with multiwalled carbon nanotubes, as positive and negative electrodes, respectively. The performance of the construct surpassed the existing 3-D printing supercapacitor devices to offer a universal strategy to form on-demand fibrous energy storage devices within wearable electronics.

The Fabrication process

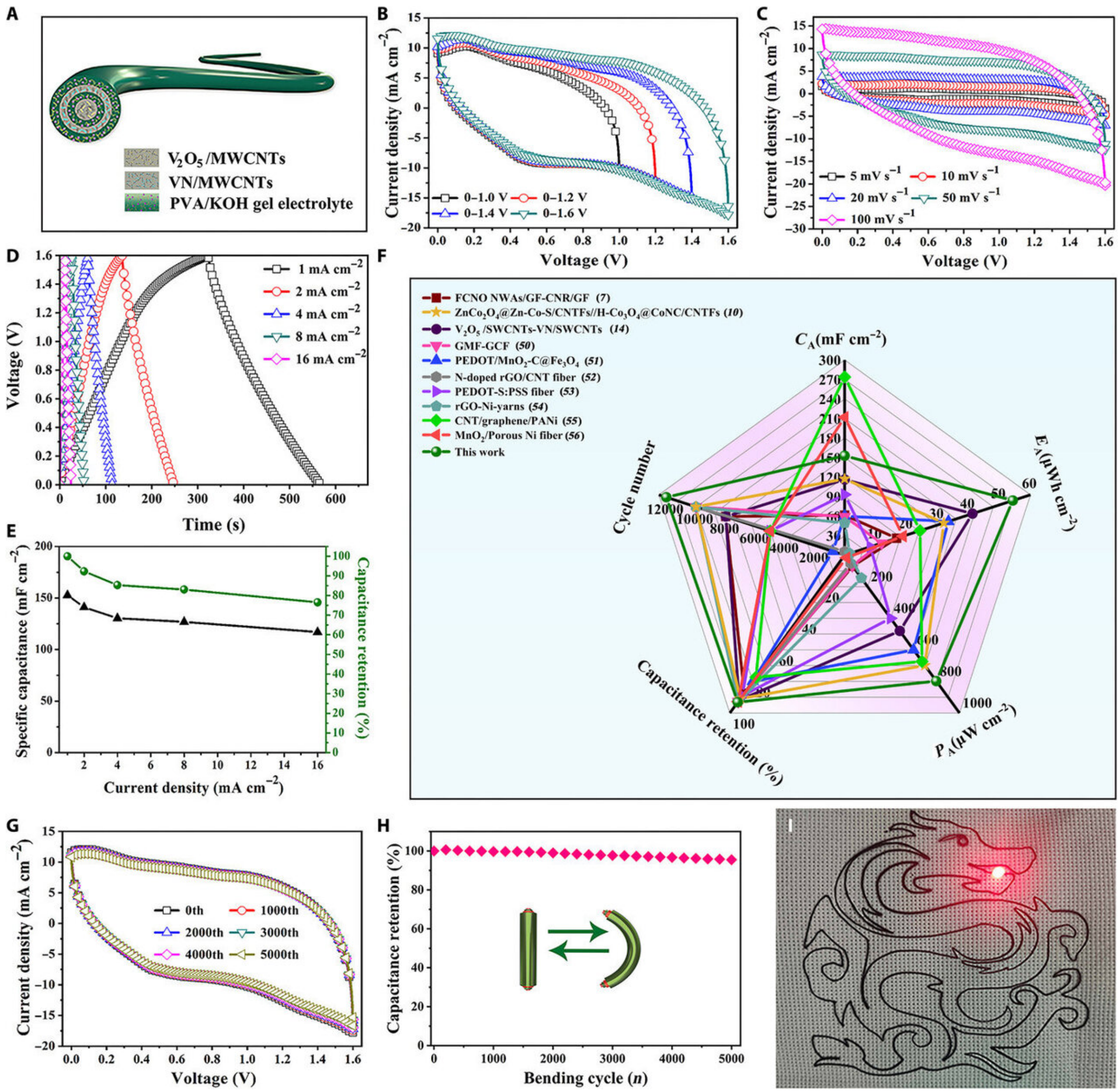
The researchers next synthesized the positive and negative electrodes to build the high-energy density FASC device. Thereafter, they uncovered the microstructure and morphology of the samples using field-emission scanning electron microscopy (FESEM) and transmission electron microscopy (TEM). They then used X-ray photoelectron spectroscopy (XPS) to survey the surface elements of the prepared samples. The team used as-printed coherent multi-inks and polyvinyl alcohol (PVA) with good rheological behavior as the 3-D printable inks to achieve the coaxial FASC device. They tuned the composition and rheology behavior of the inks for successful extrusion to maintain a self-supporting pattern. The team explained the ink behaviors with the Herschel-Bulckley model, where the values of viscosity were suitable for printing.



Structures of the electrode and 3D printing coaxial FASC device. (A to D) Schematic illustrations of the cross-sectional view of the V2O5 NW/MWCNT fiber, V2O5 NWs/MWCNTs@gel electrolyte fiber, V2O5 NWs/MWCNTs@gel electrolyte@VN NW/MWCNT fiber, and the 3D printing coaxial FASC device struts. The cross-sectional SEM images of (E) V_2O_5 NW/MWCNT fiber, (F) V_2O_5 NWs/MWCNTs@gel electrolyte fiber, (G) V_2O_5 NWs/MWCNTs@gel electrolyte@VN NW/MWCNT fiber, and (H) the 3D printing coaxial FASC device by DCMW. (I to N) The printed FASC device with different patterns. Scale bars, 50 μm (E and F), 100 μm (G and H), and 10 mm (I to N). Photo credit: (I to N) Hongyu Lu, Xi'an University of Technology. Credit: Science Advances, doi: 10.1126/sciadv.abd6978

Materials characterization and electrochemical flexible performance of the device

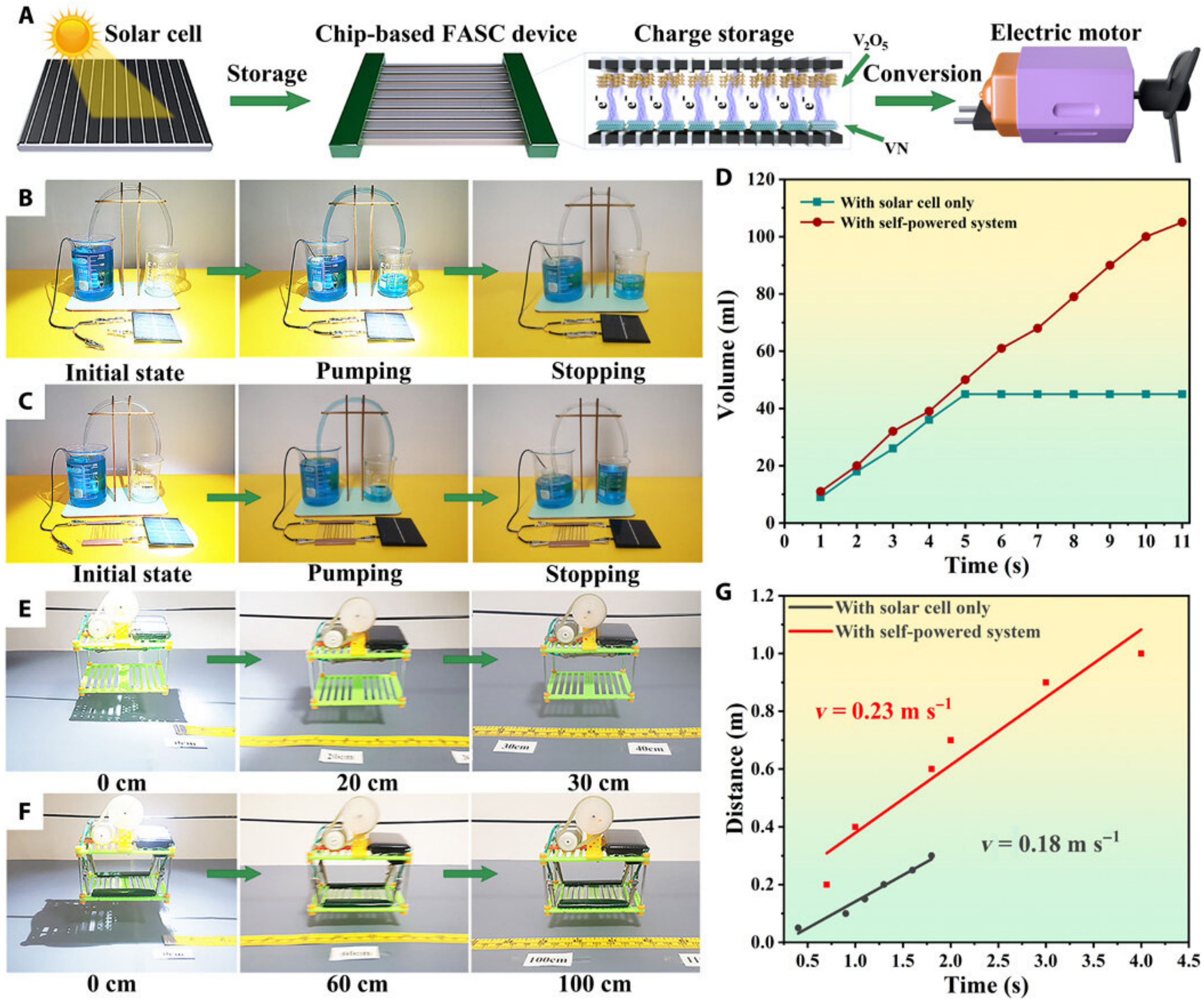
The team characterized the cross-sections of scanning electron microscopy (SEM) images of the different variants of positive and negative electrodes developed in the lab. They confirmed the phase composition and chemical states of the material ink by using X-ray powder diffraction, X-ray photoelectron spectroscopy and Raman spectra. The team observed the cross-sectional SEM image of the 3-D printing coaxial FASC device and also printed a variety of complicated patterns by 3-D printing DCMW technology to demonstrate the competence of the setup to form 3-D printed coaxial FASC devices with high accuracy and scalability. The stress-strain performance results showed excellent flexibility and mechanical strength of the printed fiber-electrodes and devices. The team observed the mesopore structures of the positive and negative electrode fibers on the basis of the pore size distribution, which benefitted the transport and diffusion of electrolyte ions during the fast charge/discharge process.



Electrochemical performance of the 3D printing coaxial FASC device. (A) Schematic diagram of the assembled device. (B) Cyclic voltammetry (CV) curves of the obtained device operated under different voltage windows. (C) CV curves of the device at different scan rates. (D) Galvanostatic charge/discharge (GCD) curves of the device at different current densities. (E) Rate capability of the device. (F) Comparison of electrochemical performance of this 3D printing coaxial FASC device with previous FASC devices (7, 10, 14, 50–56). Note to the terminology: CA, areal specific capacitance; EA, areal energy density; PA, areal power density. (G) CV curves obtained at the different bending cycles at a scan rate of 75 mV s−1. (H) Capacitance retention after 5000 cycles. (I) Photograph of a red 1.5-V LED illuminated by a fully charged 3D printed coaxial FASC device. Photo credit: (I) Hongyu Lu, Xi'an University of Technology. Credit: Science Advances, doi: 10.1126/sciadv.abd6978

Integrating the 3-D printing coaxial FASC device within a wearable device.

In order to realize the high energy density 3-D printing coaxial FASC device for a wearable device, Zhou et al. selected the accurate electrochemical performances of the positive and negative electrodes via charge matching. The as-printed coaxial FASC device embraced outstanding electrochemical performance and showed a high working voltage of 1.6 V. The team assessed the electrochemical performance of the fabricated 3-D printing coaxial device using galvanostatic charge/discharge (GCD) and electrochemical impedance spectroscopy (EIS). The results revealed the desired capacitive behavior for the as-prepared FASC device. The specific capacitance of the whole device surpassed most of the conventional fiber-shaped supercapacitors. To demonstrate the feasibility of powering the electronic devices, Zhou et al developed a fully charged 3-D printing coaxial FASC device in the shape of a dragon to illuminate a 1.5 V red light-emitting diode (LED).



Applications of the self-powered system. (A) Schematic diagram of the self-powered system of energy storage and conversion. The solar energy is converted into electrical energy and then into mechanical energy. (B) Photographs of water pumping prototype with solar cell only; less solution is obtained without extra energy. (C) Photographs of water pumping prototype with the self-powered configuration including chip-based FASC device and solar cell; more solution is obtained with energy storage. (D) Relationship between volume of the pumping solution and time of the solar cell and self-powered system, respectively. (E) Photographs of the running of a sightseeing cable car with solar cell only. The sightseeing cable car can run the short distance without extra energy storage. (F) Photographs of the running of the sightseeing cable car with the self-powered configuration including chip-based FASC device and solar cell. The sightseeing cable car can run the long distance with energy storage, demonstrating longer durability. (G) Relationship between the running distance and time of the sightseeing cable car with self-powered system and solar cell only, respectively. The running speed of the sightseeing cable car with self-powered system is faster than that with solar cell only. Photo credit: (B, C, E, and F) Jingxin Zhao, University of Macau. Credit: Science Advances, doi: 10.1126/sciadv.abd6978

Constructing a self-powered and self-moving system for energy storage and conversion

The scientists then integrated the FASC devices with a solar cell and electric motor to realize a self-powered system to convert solar energy into electric energy and mechanical energy. The as-fabricated 3-D printing coaxial FASC device provided power to the pressure sensor in the setup based on bioinspired multiscale structured polydimethylsiloxane (PDMS) and polypyrolle stamps due to the existence of the multiscale architecture. The team did not observe performance degradation after 600 loading/unloading cycles to demonstrate the excellent cycle stability of the device. The all-in-one coaxial solid-state FASC device with high energy density therefore proved a prospective candidate across the new fields of artificial intelligence, robotics and sensing.

In this way, Jingxin Zhao and colleagues developed a 3-D printing direct coherent multi-ink writing technology to fabricate an all-in-one coaxial solid-state FASC device with an ultrahigh areal energy or power density, with multi-inks. The compact structure of the printed coaxial FASC device embraced splendid flexibility and mechanical stability performance that was superior to traditional architecture asymmetric supercapacitors. The 3-D printing coaxial FASC devices served as on-demand energy storage units to drive pinwheels, pumping prototypes, electric cars, and pressure sensors with improved performance. The results offer a highly versatile solution to design high-performance, on-demand, fiber-based energy storage devices for advanced wearable applications.

More information: Zhao J. et al. Direct coherent multi-ink printing of fabric supercapacitors, Science Advances, DOI: 10.1126/sciadv.abd6978

Shi P. et al. Design of amorphous manganese oxide@multiwalled carbon nanotube fiber for robust solid-state supercapacitor, ACS Nano, doi.org/10.1021/acsnano.6b06357

Lee A. et al. 3-D bioprinting of collagen to rebuild components of the human heart. Science, DOI: 10.1126/science.aav9051

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