

REVIEW ARTICLE OPEN



Green flexible electronics based on starch

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Flexible electronics (FEs) with excellent flexibility or foldability may find widespread applications in the wearable devices, artificial intelligence (AI), Internet of Things (IoT), and other areas. However, the widely utilization may also bring the concerning for the fast accumulation of electronic waste. Green FEs with good degradability might supply a way to overcome this problem. Starch, as one of the most abundant natural polymers, has been exhibiting great potentials in the development of environmental-friendly FEs due to its inexpensiveness, good processability, and biodegradability. Lots of remarks were made this field but no summary was found. In this review, we discussed the preparation and applications of starch-based FEs, highlighting the role played by the starch in such FEs and the impacts on the properties. Finally, the challenge was discussed and the outlook for the further development was also presented.

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INTRODUCTION

More than 100 years have passed since human walked from vacuum tubes to transistors. Nowadays, electronic devices are transforming from solid, durable, single-shape styles to wearable, versatile, high-performance, and multifunctional components, among which the flexible electronics (FEs) have already sprung up and gained extensive attentions^{1–4}. The appealing applications of FEs may at least include personalized mobile devices, health-care systems, human-machine interfaces, soft robots, electronic skin, artificial intelligence (AI), and Internet of Things (IoT)^{5–8}. To fabricate FEs, the soft, stretchable, foldable substrates are highly desired. In general protocols, electro-conducting materials including metals, conducting polymers, or carbon-based materials are assembled on the flexible substrates, which involves polymer films such as polyethylene terephthalate (PET)⁹, polyimide (PI)^{10,11}, hydrogels¹², textiles¹³, and elastomers such as polydimethylsiloxane (PDMS)^{14,15}. Nevertheless, these substrates are usually non-degradable, which may result in accumulation of electronic waste (e-waste). In estimation, the globe production of e-waste reached to 53.6 million tons by 2020, an average of 7.3 kg of one person. And this number might increase to 74.7 million tons by 2030¹⁶. In 2019, only 17.4% of the e-waste was recycled due to the tedious process and imperfect policy. Green FEs with good degradability may supply a way to fundamentally solve the problem.

In recent years, a variety of environment-friendly, bio-degradable, and low-cost electronic products have emerged^{17,18}, which were made of materials from nature, including cellulose^{19–24}, lignin^{25–29}, proteins^{30–32}, and starch^{33–35}. These materials from nature have intrinsic advantages of abundant sources, low cost, good degradability, acceptable biocompatibility, fine accessibility for chemical modification comparing with synthesized polymers. They can be safely and bio-compatibly used as implantable devices for human healthcare and medical diagnose³⁶. Furthermore, their utilization may be free of e-waste production to satisfy the requirements of the e-waste reduction.

Among the reported sustainable materials used in FEs, starch have advantages on the acceptable solubility in water, good property of film forming and relative low cost comparing with

cellulose, lignin and proteins. Therefore, a lot of efforts have been put into the developments of FEs based on starch^{37–41}, in which the starch usually appeared as films or gels. The starch films were often used as soft and transparent substrates because of the intrinsic good film-forming property of starch. The conductive materials can be easily coated or printed on the starch film to fabricate FEs. On contrary, the starch hydrogel can be feasibly processed to various shapes with injection or 3D printing, which may further carbonize into conductive materials with stereoscopic structures. The water containing system of the hydrogel may also benefit for their biomedical application.

Though the starch films and gels have their own features, both were versatile in the exploration of FEs, such as flexible conductive electrodes, sensors, supercapacitors, transistors, and resistive switching memory devices (Fig. 1). However, to the best of our knowledge, no summary on this exciting area was found. Therefore, here in this review, the fundamentals and advantage of the starch used for FEs were firstly discussed. Subsequently, the starch-based devices were divided into two categories and successively summarized, i.e., FEs based on starch films vs FEs based on starch gels. The strategies of the preparation were introduced and the role of the starch played in these FEs was highlighted. The challenges and future prospects were finally addressed to end the review.

Starch is a natural-derived polysaccharide comprising of glucose monomers, which can be abundantly harvested from the roots, stems, and seeds of the rice, corns, wheat, cassavas, potatoes, and other crops. According to the molecular chain structure, the starch can be divided into amylose and amylopectin^{42–44}. In the natural starch, amylose accounts for 20~26%, and the rest are amylopectin. The amylose has a linear monomer at chain with only α -1,4 linkages thus possessing good extensibility in solution and can easily associate with some polar organic compounds by hydrogen bond. The amylopectin is the branch form with α -1,4 linkages in the backbone and α -1,6 linkages at the branched points, which usually has much higher molecular weight⁴⁵. In addition, starch molecules contain lots of hydroxyl groups that are suitable for esterification⁴⁶, etherification⁴⁷, grafting⁴⁸, cross-linking⁴⁹, and

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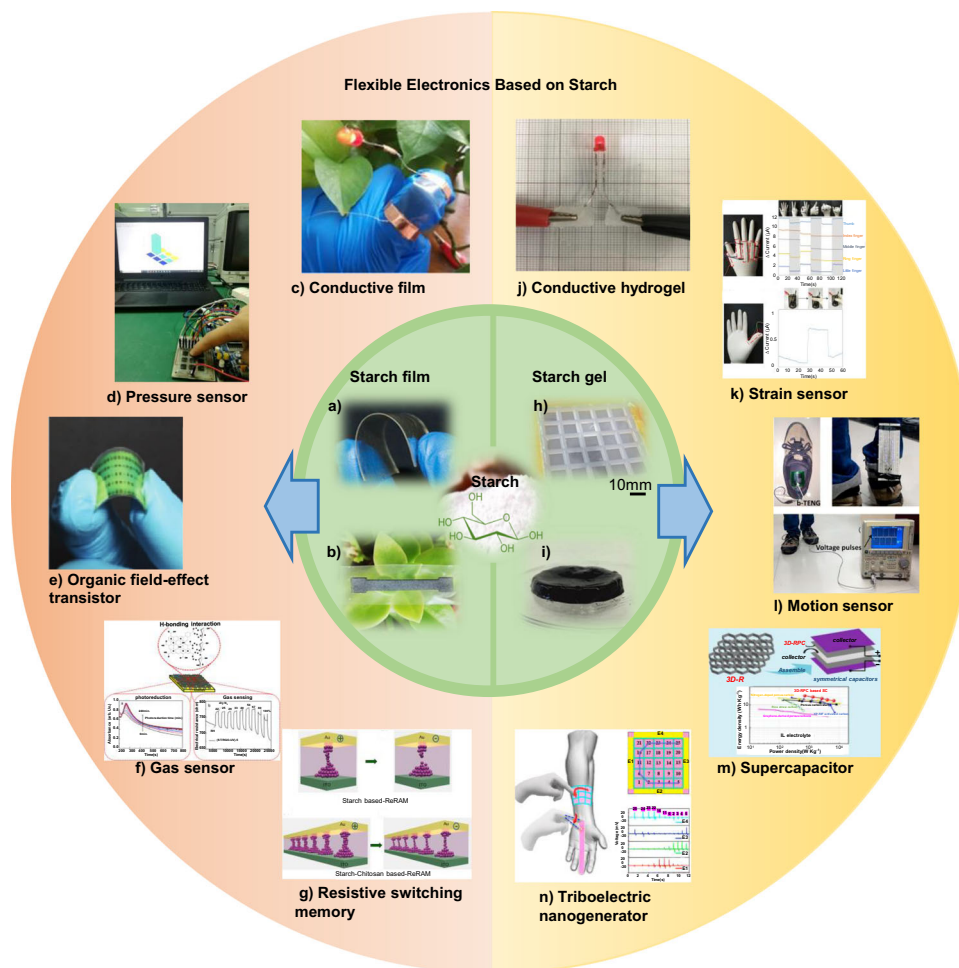


Fig. 1 Summary of starch-based FEs. Left, device on starch films: Starch film (a). Reproduced with permission⁸⁶. Copyright 2019, Springer. Starch film (b). Reproduced with permission³⁷. Copyright 2019, American Chemical Society. c Conductive film. Reproduced with permission³⁸. Copyright 2018, American Chemical Society. d Pressure sensor. Reproduced with permission³⁷. Copyright 2019, American Chemical Society. e Organic field-effect transistor. Reproduced with permission³³. Copyright 2017, Wiley. f Gas sensor. Reproduced with permission⁹⁸. Copyright 2020, American Chemical Society. g Resistive switching memory. Reproduced with permission³⁵. Copyright 2016, American Chemical Society. Right, device on starch gels: Starch gel (h). Reproduced with permission³⁹. Copyright 2018, American Chemical Society. Starch gel (i). reproduced with permission¹⁴⁷. Copyright 2019, Elsevier. j Conductive hydrogel. Reproduced with permission⁴⁵. Copyright 2019, American Chemical Society. k Strain sensor. Reproduced with permission¹⁵¹. Copyright 2019, Elsevier. l Motion sensor. Reproduced with permission⁴⁰. Copyright 2019, American Chemical Society. m Supercapacitor. Reproduced with permission¹⁵⁸. Copyright 2019, American Chemical Society. n Triboelectric nanogenerator. Reproduced with permission³⁹. Copyright 2018, American Chemical Society.

other chemical modification^{50,51}. Moreover, starch has outstanding environmental degradability and good biocompatibility. These characteristics make it an important candidate as raw material for fabricating green FEs.

Starch, is the most widely used biodegradable materials for now. In Europe, 50% of the produced starch was used for non-food applications⁵², such as the composition in adhesives and paper binders, textiles, chemical production, the feedstock for fermentation and other industrial products⁵³. In addition, starch has physical, chemical, and functional properties, including good water solubility, gelatinization, high temperature adhesion behavior, and easy modification. The utilization of starch as a platform for building FEs at least has following advantages: (1) Starch is one of the most abundant renewable materials with extremely low cost. (2) Starch is degradable in water and soil without toxic residues thus being regarded as environmentally friendly materials. (3) Starch is biocompatible and suitable for implantable electronics. (4) Starch is easily transformed into flexible, foldable films and gels. (5) Starch is light in weight. Comparing with other biomass materials for FEs, the starch still has some advantage.

For instant, it shows better solubility than cellulose and it holds lower cost than proteins^{53,54}.

FES BASED ON STARCH FILMS

Starch is easy to be transformed into a film through heating in the presence of water followed by casting and drying. However, the starch films prepared by pure natural starch are usually fragile with low transparence. They are usually reinforced by compositing with other polymers, adding plasticizers, or modification through physical or chemical strategies^{38,55,56}. The modified starch films may obtain acceptable mechanical property^{57,58}, good flexibility⁴⁹, or other features such as electrical conductivity⁵⁹, making them more favorable to be utilized as soft substrates or the constituents of FEs. The data of FEs based on starch films were listed in Table 1.

Conductive films derived from starch

Conductive films can be easily casted with starch when electroconductive materials are encapsulated. The most frequently used conducting components include graphene,

Table 1. Summary of FEs based on starch films.

Device Type	Materials	Performance	Function of starch	Ref
Conductive film	Starch, graphene	$9.7 \times 10^{-4} \text{ S cm}^{-1}$	Modification of graphene	48
Conductive film	Starch, SWCNT	$10^{-3} \text{ S cm}^{-1}$	Substrate	61
Conductive film	Starch nanocrystals, GO	$65.8 \mu\text{S m}^{-1}$	Dispersing agent	65
Conductive film	Starch, SCNT-PG-PEDOT	$46 \Omega \text{ sq}^{-1}$	Substrate	38
Conductive film	Starch, ionic liquid	$>10^{-3} \text{ S cm}^{-1}$	Substrate	59
Conductive film	Starch, graphene	$3.9 \times 10^{-4} \text{ S cm}^{-1}$	Filler	66
Conductive film	Mater-Bi, graphene	$\sim 10 \Omega \text{ sq}^{-1}$	Dispersing agent	70
Conductive film	Starch, AuNP	$<1.0 \Omega \text{ sq}^{-1}$	Reducing and stabilizing agents	71
Strain sensor	Starch, egg white, Ag	$<1.0 \Omega \text{ sq}^{-1}$,	Substrate	86
Gas sensor	Starch, CNT	Sensitivity enhanced	Functionalization of CNT	97
Gas sensor	Potato starch, graphite flakes	10–1000 ppm	Functionalization of graphite	98
Chemical sensor	Starch, GO, AuNP	1.5 to $22 \mu\text{mol L}^{-1}$	Substrate	87
Multifunctional sensor	Starch, porous carbon	Strain (GF = 134.2), temperature (25–90°C), pressure (0–250 kPa)	Substrate	37
Supercapacitor	Starch, NaClO_4 , glutaraldehyde	ESW~2.4 V	Electrolyte	106
Supercapacitor	Starch, Ni quantum dots	Specific capacitance 1120 F g^{-1}	Assistant	107
Nanogenerator	Starch, lignin	Power density 173.5 nW cm^{-2}	Electrode	119
Nanogenerator	Starch	Output voltage 11.2 V	Electrode	120
Nanogenerator	Starch, cellulose	Output voltage 60–300 mV	Electrode	121
Nanogenerator	Starch	Output voltage 1.2 V	Electrode	122
Nanogenerator	PDMS, starch	Output voltage ~560 V	Electrode	40
Transistors	Starch, glycerol	On/off ratio 2.6×10^6	Ion-based gate dielectric	128
Organic transistor	Starch, PVA, Ag	On/off ratio 6.9×10^4 – 4.9×10^5	Substrate	33
Memristor	Starch, chitosan, Au, ITO	On/off current ratio $\sim 10^3$	Active layer	35
Electrolyte membranes	Starch, glycerol, ionic liquid, urea	Ionic conductivity $6.2 \times 10^{-4} \text{ S cm}^{-1}$	Proton exchange membrane	135

conducting polymers, ionic liquid, metals, and their alloys⁶⁰. Zheng et al.⁴⁸ grafted starch molecules on the surface of graphene nanosheets (GN) in the presence of hydrazine hydrate. The conducting films were then fabricated with GN-starch filled plasticized starch, which shown conductivity up to $9.7 \times 10^{-4} \text{ S cm}^{-1}$ when the content of GN-starch was only 1.774 wt%. The film also possessed excellent mechanical properties and moisture resistance, because the GN was well dispersed with the assistance of starch. Prusty et al.⁶¹ prepared nanocomposites of functionalized single-walled carbon nanotubes (f-SWCNTs) and polyacrylonitrile-co-starch (PAN-co-starch) using in-situ polymerization technique. The nanocomposites have excellent electrical conductivity up to $10^{-3} \text{ S cm}^{-1}$.

The starch nanocrystals (SNCs) can be obtained after removing the amorphous zone by the treatment of enzyme, sulfuric acid, or hydrochloric acid^{62,63}. The utilization of the starch nanocrystals to prepare composites may not only improve the mechanical properties of materials, but also increase the biodegradability^{63,64}. Zhu and co-workers⁶⁵ extracted plate-like SNCs from natural waxy corns, which were then used to improve the dispersion of graphene in the composite film based on soybean protein (Fig. 2a). The composite conductive film shown a good conductivity of $65.8 \mu\text{S m}^{-1}$, enhanced mechanical strength, reduced water vapor and oxygen permeability as well as fine solvent resistance.

The conductive films with good transparency can also be prepared from starch. Miao and co-workers³⁸ spray-coated the carbon nanotube (CNT), pristine graphene (PG), and poly(3,4-ethylenedioxythiophene) (PEDOT) on the composite film of starch and chitosan to fabricate conducting film (Fig. 2b), which shown transmittance of 83.5% at 550 nm and low sheet resistance of $46 \Omega \text{ sq}^{-1}$ as well as good mechanical performance. The conducting film worked as a transparent flexible electrode. Furthermore, it

can be rapidly degraded in lysozyme solution at room temperature indicated it is a green electronic device. Similar transparent film recently developed with graphene and potato starch, which achieved conductivity of $3.9 \times 10^{-4} \text{ S cm}^{-1}$ with good moisture barrier properties⁶⁶.

The most reported protocol to prepare starch films involved dissolving in water, gelatinizing and casting^{67,68}. Plasticizers such as polyols (glycerol, ethylene glycol) or citric acid have to be added to reinforced the films^{49,55,69}. Different from traditional-used plasticizers, ionic liquid (1-ethyl-3-methylimidazoliumacetate, $[\text{C}_2\text{mim}][\text{OAc}]$) was employed as a functional plasticizer by Zhang and co-worker. to prepare electroconductive and transparent film (Fig. 2c)⁵⁹. The film had good electrical conductivity ($>10^{-3} \text{ S cm}^{-1}$) and was straightforwardly processed at moderate temperature (55–65 °C).

Except for solution casting as films, starch was used as a stabilizer in the preparation of conductive ink, which was then applied to fabricate conductive films. Cataldi et al.⁷⁰ dispersed commercial thermoplastic starch-based polymers (Mater-Bi, blends of thermoplastic starch and aliphatic polyester) and graphene nanosheets in an organic solvent to generate conductive inks, which was sprayed on pure cellulose substrate (Fig. 2d), and then hot pressed to form flexible conductive film (Fig. 2e). The sheet resistance reached as low as $10 \Omega \text{ sq}^{-1}$ by regulating of the content of the graphene nanoplatelets. It was noted that the composite film maintained its mechanical and electroconductive performance even under many severe folding events, which is significant for flexible conductors. In the preparation of printable conductive ink, starch can also work as a reducing agent for metal ions. Bacalzo et al.⁷¹ synthesized an aqueous gold nanoparticle (AuNP) ink suitable for inkjet printing using hydrolyzed starch as reducing agent and stabilizer (Fig. 2f). The AuNP ink can be

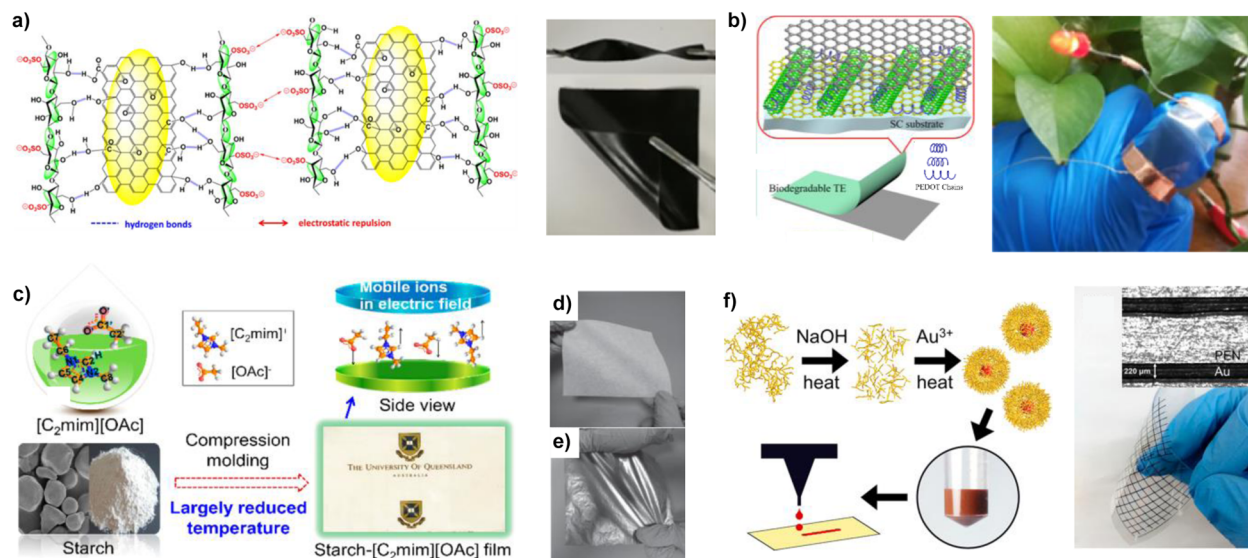


Fig. 2 Conductive films derived from starch. **a** Mechanism of the starch-aided dispersion of graphene and a photograph of the corresponding conductive film. Reproduced with permission⁶⁵. Copyright 2017, American Chemical Society. **b** Structural illustration and photograph of flexible conductive film with CNT-PG-PEDOT on starch and chitosan composite. Reproduced with permission³⁸. Copyright 2018, American Chemical Society. **c** Starch-based conductive films prepared with ionic liquid as plasticizer. Reproduced with permission⁵⁹. Copyright 2017, American Chemical Society. **d** Photograph of the porous cellulose fiber substrate and **e** the paper-like conductor after impregnating with inks of Mater-Bi starch-based polymer and graphene. Reproduced with permission⁷⁰. Copyright 2015, Wiley. **f** Preparation process and image of the conductive film with inkjet printing of AuNP ink using starch as reduce agent. Reproduced with permission⁷¹. Copyright 2018, American Chemical Society.

printed and sintered at low temperature ($<200^{\circ}\text{C}$) to generate conductive film with resistance as low as $1.0\ \Omega\ \text{sq}^{-1}$. In this strategy, the starch solution was heated and hydrolyzed in NaOH solution, and then activated to reduce Au^{3+} in HAuCl_4 solution to Au^0 . Some starch molecules were hydrolyzed to stabilize the nanoparticles and control their further growth. This water-based ink can be used to print conductive patterns on a variety of substrates, demonstrating the versatile possibility of application in FE devices development.

Sensing elements on starch films

Flexible sensing devices have important applications in the fields of human health monitoring, human-machine interface and wearable devices, which have attracted extensive attentions in recent year^{13,72,73}. According to the application of detection, the sensing devices can be divided into physical sensors^{74–76}, chemical sensors^{77–79}, and biological sensors^{80–82}. The starch have been used to develop degradable or recyclable flexible sensors because it is biocompatible and reproducible.

Strain sensor. Strain sensor is a device that transforms deformation into electrical signal, which is widely used in wearable devices^{75,83–85}. Liu et al.⁸⁶ demonstrated a high-performance transparent strain sensor using starch to fabricate conductive matrix. The egg white was used as the sacrificial layer to support the conducting networks of silver by electro-plating. The starch solution was then dip-coated on the surface of the silver mesh and generated self-supported starch film with silver networks embedded (Fig. 3a), which shows low electro-resistivity ($<1.0\ \Omega\ \text{sq}^{-1}$) and high transparency (82%) (Fig. 3b). As shown in Fig. 3c, the resistance of the strain sensor changes following multiple bending cycles, which can be used to detect movements of human joints. The starch can be easily degraded and removed by water to yield an independent Ag network, indicating that the devices is recyclable.

Chemical sensor. Chemical sensors are used to detect chemicals in liquid or gas state, which have potentials in environment

monitoring, chemical diagnose, warning of hazardous gas. Jodar et al.⁸⁷ modified glassy carbon electrodes with RGO, gold nanoparticles (GNPs), and potato starch (PS) to produce a stable RGO-GNPs-PS conductive film as an electrochemical sensor for detection of estriol (Fig. 3d), which was a pollutant found in environment and aquatic organisms. The starch had good film-forming property and chemical stability, which was demonstrated in good supportability and permeability, as well as to compensate for the surface roughness of the electrode. The linear response of estriol was found in the range of $1.5\text{--}22\ \mu\text{mol L}^{-1}$, and the detection limit was $0.48\ \mu\text{mol L}^{-1}$. The RGO-GNPs-PS also effectively recycled 92.1–106% of estriol in natural water samples and synthetic urine. Similar strategies were used to fabricate electrochemical sensors for the detection of various chemicals^{88–92}, in which the starch played an important role such as the substrates.

Gas sensor. The gas sensor is a device that detects various gas such as different organic vapors. It is also known as electronic nose^{79,93–95}. Starch can be used to tune the nanostructure of gas sensor to enhance its sensitivity and selectivity⁹⁶. Kumar et al.⁹⁷ investigated the use of amylose and amylopectin functionalized carbon nanotubes (CNT) to fabricate electronic noses on micro-electrodes by spraying them layer by layer to detect vapors of water, methanol and toluene. As shown in Fig. 4a, they adsorbed on the surface of carbon nanotubes in helical or random conformation, respectively, which changed the junction gap of CNT. Interestingly, amylose increased the sensitivity of the CNT sensor to water, while amylopectin increased the sensitivity to toluene (Fig. 4b). In another example, Peregrino et al.⁹⁸ prepared gas sensor by assembling starch and graphene oxide (GO) layer by layer on a quartz substrate (Fig. 4c). They found at least three important roles that the starch was played. At first, hydrogen bondings were established between $-\text{OH}$ groups in starch and $-\text{C}-\text{O}-\text{C}-$, and $=\text{C}=\text{O}$ from GO, which improved the efficiency of film assembling. Secondly, the starch provided additional electron density during photoreduction of GO, making the reduction faster and more efficient. Thirdly, the sensitivity of the device to humidity was improved since the starch is hygroscopic.

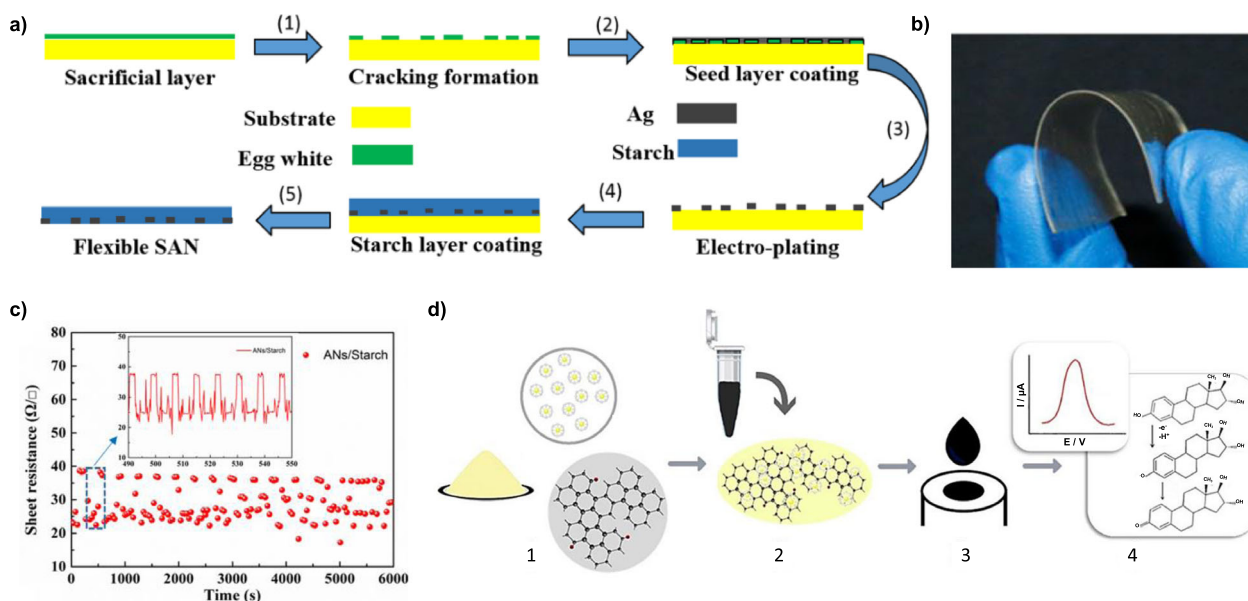


Fig. 3 Flexible strain and chemical sensors based on starch films. **a** Fabrication processes and **b** flexibility demonstration as well as **c** sheet resistance corresponding to bending times of a sensor based on starch-Ag networks (SANs). Reproduced with permission⁸⁶. Copyright 2019, Springer. **d** Preparation protocol of the chemical sensor that detects estriol from RGO-GNPs-PS. Reproduced with permission⁸⁷. Copyright 2017, Springer.

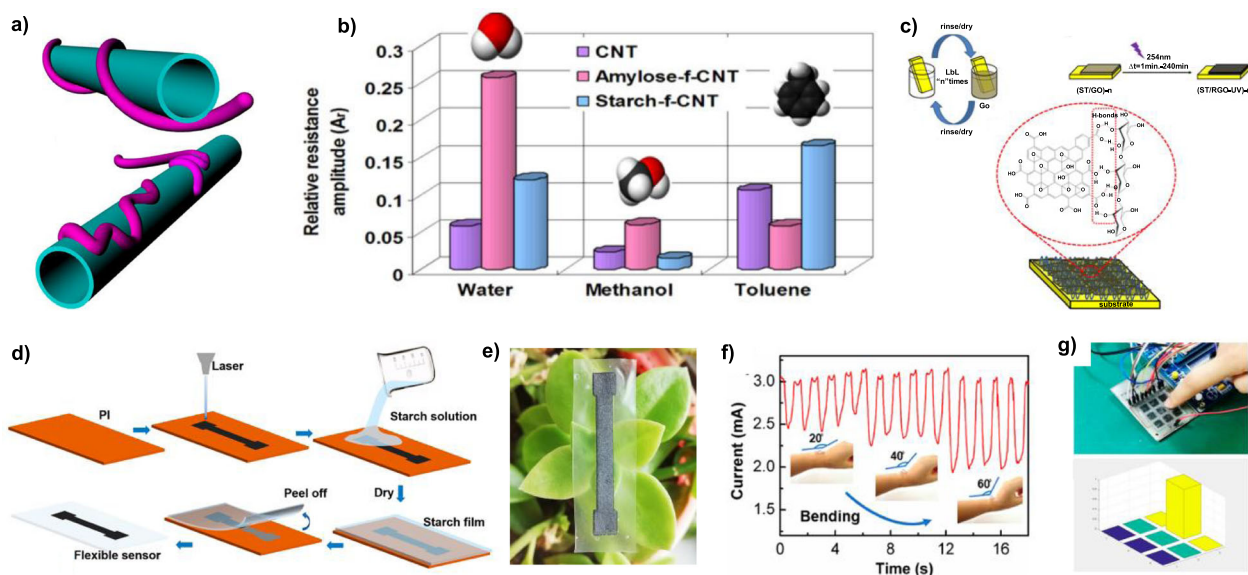


Fig. 4 Flexible gas and multifunctional sensors based on starch films. **a** Possible conformation of amylose-f-CNT and starch-f-CNT sensors and **b** their selectivity for different vapors. Reproduced with permission⁹⁷. Copyright 2012, Elsevier. **c** Illustration of the layer-by-layer (LbL) deposition, photochemical reduction, and hydrogen bonding of the starch-GO sensor on quartz substrate. Reproduced with permission⁹⁸. Copyright 2020, American Chemical Society. **d** The preparation procedure and **e** photograph of a multimodal flexible sensor on starch film. **f** Real-time current signal for monitoring of wrist bending with different angles. **g** The sensor matrix for detection of spatial pressure. Reproduced with permission³⁷. Copyright 2020, American Chemical Society.

Therefore, it is convincing that the starch may find more applications in fabricating such intelligent device.

Multifunctional sensor. The preparation of FEs with low cost and high precision detection of various stimuli are the significant challenges^{12,99,100}. In our previous work³⁷, laser-induced porous carbon was transferred to the starch film producing a flexible multimodal sensor (Fig. 4d, e). The strain, humidity, temperature, and pressure can be detected with one device, thus decreasing the price of sensor matrix. The sensor can detect strain with gauge factor (GF) of 134.2, response time of approximately 130 ms, and

good durability more than 1000 cycles of bending-unbending, which can be used to monitor human motions including bending of fingers, wrists (Fig. 4f) and knees. The temperature change between 30 and 50 °C and pressure in the range of 0–250 kPa were also detected by the sensor. The response time for pressure detection was ~0.15 s. Interestingly, the sensors in matrix pattern were easily fabricated with this strategy, which precisely detected spatial pressure (Fig. 4g). Furthermore, multiple stimuli can be distinguished with the sensor, such as pressure coupled with temperature change. More importantly, the sensor can be degraded in water thus being free of e-waste, so it is a typical green electronic device.

Supercapacitor on starch films

Traditional energy sources cannot fulfill the dramatical increment of energy demands in nowadays, thus the alternate and renewable energy source attracted widely attentions. The electrolyte is one of the most important parts of renewable energy device. Polymers including polyethylene oxide (PEO), poly (vinylidene fluoride) (PVDF), and poly (methylmethacrylate) (PMMA) were intensively investigated as host of salt complex electrolytes in previous research^{101–105}. Recently, people found the starch can meet the 3Es requirement of electrolytes that are (a) environment friendly, (b) economical and (c) easy to mould in desired shape and size. For instance, Chauhan et al.¹⁰⁶ prepared a highly transparent film with conductivity up to $10^{-2} \text{ S cm}^{-1}$ using composite of corn starch and NaClO_4 after cross-linking with glutaraldehyde. The equivalent series resistance of the sample with thickness of 0.8 mm was $\sim 6.252 \Omega$, the electrochemical stability window (ESW) was $\sim 2.4 \text{ V}$ and the ion relaxation time was $\sim 65 \mu\text{s}$. Chen and co-workers¹⁰⁷ synthesized ultrathin carbon nanosheet-supported Ni quantum dot hybrids (C-Ni-QDs) through hydrothermal method followed by annealing process with the assistance of starch. The C-Ni-QDs was used to assemble a supercapacitor, which shown a high specific capacitance up to 1120 F g^{-1} at 2 A g^{-1} and a capacitance retention of 97% after 2000 cycles. These results shown that the starch is a promising carbon sources for supercapacitors that is inexpensive and abundant.

Nanogenerator on starch films

Nanogenerators based on triboelectric, piezoelectric, and electrostatic induction effects have been proposed to construct self-actuating systems that produce electricity from various mechanical energies, including human motion^{108–110}, vibration, and wind^{111,112}. The flexible nanogenerators can provide a flexible and portable energy collection system or work as self-powered sensors, which has recently become a hot research topic^{9,113–116}.

Triboelectric nanogenerators (TENGs) are made of two materials with different triboelectric properties. The friction between the two films generate equal but opposite charges on both sides due to nanoscale surface roughness, while the friction potential layer generated in the interface region drives the flow of electrons in the external load^{117,118}. Bao et al.¹¹⁹ developed TENGs based on lignin-starch nanocomposites (Fig. 5a), in which the starch improved the uniformity of the film. When the ratio of lignin to starch in the composite film was 3:7, the highest output was reached. The average short-circuit current was 3.96 nA cm^{-2} , and the open-circuit voltage was 1.04 V cm^{-2} . Zhu et al.¹²⁰ assembled TENGs based on a starch film, using the coupling effect between human skin and starch film to develop a self-powered sensor for detecting human perspiration (Fig. 5b). Vela and co-workers¹²¹ described a starch-cellulose-based TENG using potato starch, and sandpaper as the substrate of microstructure. They found that the output voltage depended on the thickness (50–200 μm) of the film. The electric output of 4 cm^2 TENG reached up to 300 mV in the presence of the starch film. Further research by Vela prepared a low-cost TENG using a starch film with micro-structure as a triboelectric dielectric layer (Fig. 5c)¹²². The 0.5% of CaCl_2 was added into the film to improve the voltage output from 0.4 to 1.2 V. Sarkar and co-workers⁴⁰ developed TENGs using thermo-plastic starch film as a positive electrode and PDMS as negative electrode (Fig. 5d). This bilayer TENG (b-TENG) can generated open-circuit output voltage up to $\sim 560 \text{ V}$ and output current density of around $\sim 120 \text{ mA m}^{-2}$, which can light more than 100 LEDs. In addition, the TENG device can work as a self-powered pedometer for walking and a gait analysis sensor for health evaluation (Fig. 5e, f). The good performance of the devices benefited from the surface morphology of the starch film, which

may open a window for the low cost and environmental-friendly self-powered electronic devices.

Organic transistor on starch films

Flexible organic field-effect transistor (OFET) devices have attracted much attention as a display technology^{123–127}. Intrinsically flexible OFET has significant advantages compared with inorganic thin film transistors on accelerate technology update, simple production process and low cost. Shao et al.¹²⁸ used starch as an ion-base gate dielectric for oxide thin film transistors (Fig. 6a). It was found that the transistor performance is closely related to the specific capacitance and ionic conductivity of starch (Fig. 6b). Higher on/off ratio and field mobility can be achieved by glycerol incorporated starch. The results demonstrated the potential application of starch in thin film transistors. In Jeong's work³³, a flexible OFET was fabricated on starch-PVA film by a vapor-deposited method (Fig. 6c). The starch film has good transparency (Fig. 6d), remarkable mechanical strength, and good stability in non-polar solvent. The calculated field-effect mobility was $0.013\text{--}0.37 \text{ cm}^2 (\text{Vs})^{-1}$ with on/off ratio of $6.9 \times 10^4\text{--}4.9 \times 10^5$ (Fig. 6e), which were comparable to that of other flexible OFETs. Interestingly, the device on the film can be quickly degraded in fishbowl water by the fungi inside, suggesting it is eco-friendly biodegradable FEs with low cost.

Other electronics on starch films

Resistive switching memories (ReRAMs) are the major candidates for replacement of the state-of-the-art memory devices in the future due to their good scalability, ultrafast write and read access, and low power consumption¹²⁹. The ReRAM using polymer as the active layer has the advantages of good flexibility, low cost, good processability and fine compatibility with various substrate. Biopolymers such as chitosan¹³⁰ and cellulose¹³¹ have been considered in the electronic devices of synapses^{28,131,132}. Due to the presence of loosely bound water molecules in the crystal network, the potato starch has reasonable ionic conductivity^{133,134}. Raeis-Hosseini et al.³⁵ demonstrated a non-volatile, flexible and transparent ReRAMs device using potato starch and chitosan as active layer (Fig. 7a). The device was prepared with Au/starch-chitosan/ITO on PET flexible substrate by spin coating and thermal evaporation. The inset images in Fig. 7a shows a magnified optical image of the storage array on the flexible substrate. The resistance behavior of the device can be controlled effectively by the content of starch and chitosan as well as the chemical composition of starch (amylose and amylopectin). Both Au/starch/ITO/PET and Au/starch-chitosan/ITO/PET device shown good set/reset performance (Fig. 7b, c). Furthermore, the progressive setting/resetting behavior of this storage devices makes them potential in the application of neuromorphic devices such as simulation-based synaptic electronics.

Fuel cells and redox flow batteries provide considerable energy and power density that are more sustainable alternatives to lithium batteries. However, the perfluorosulfonic acid copolymer proton exchange membranes in these cells are still an important environmental problem. In Alday's work¹³⁵, biopolymer electrolyte membranes (Bio-PEMs) were synthesized from starch, cellulose, and chitosan. The starch-based Bio-PEMs were doped with ionic liquid, glycerol, or urea (Fig. 7d), which reached the highest ionic conductivity of $6.2 \times 10^{-4} \text{ S cm}^{-1}$ through a symmetrical cell test (Fig. 7e). These Bio-PEMs are promising alternatives for conventional copolymer membranes in sustainable electrochemical systems and biodegradable devices.

FES BASED ON STARCH GELS

Gels derived from biopolymers have attracted increasing attentions as a result of their good biocompatibility, innate

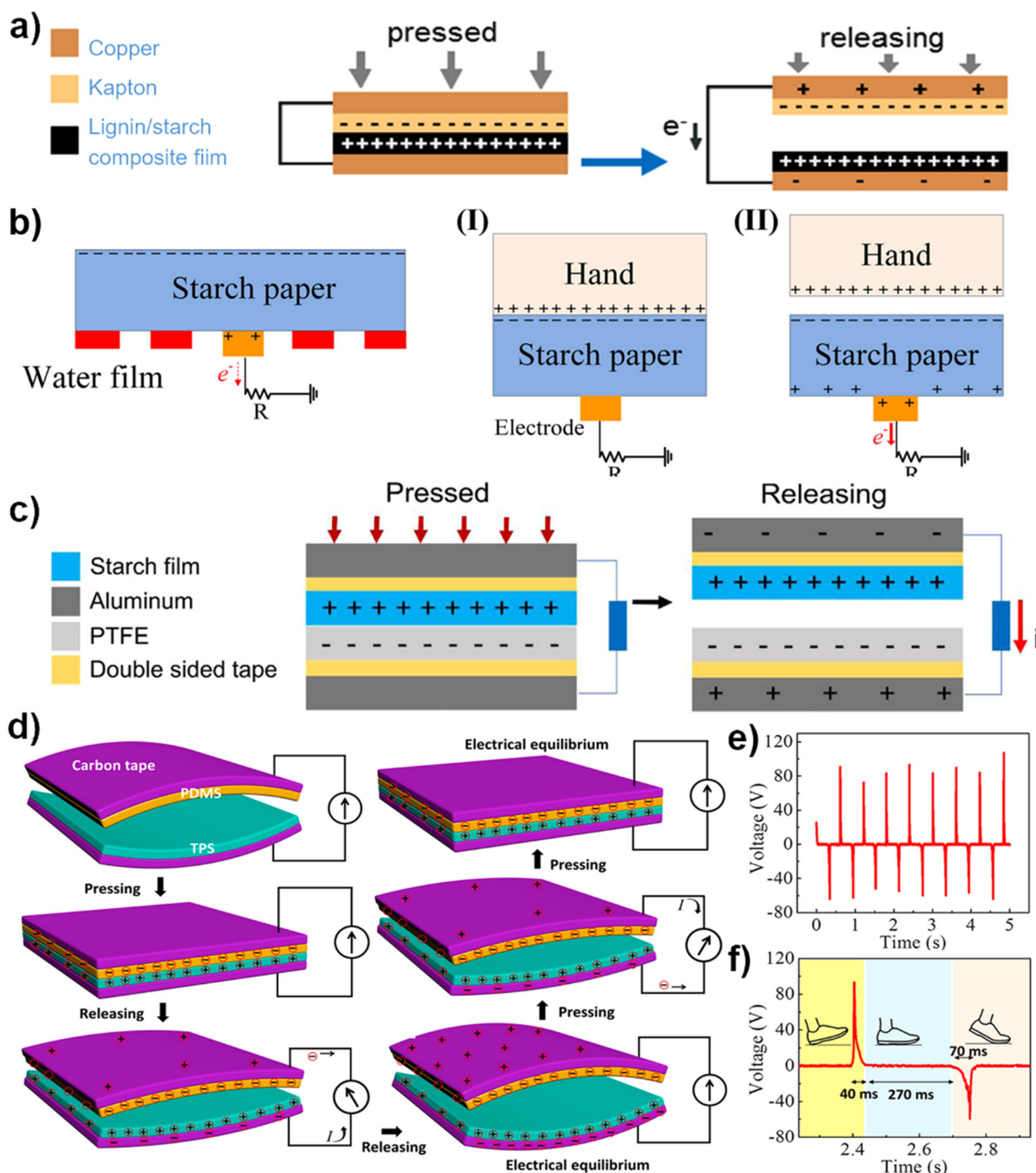


Fig. 5 TENGs based on starch films. **a** Mechanism of lignin-starch nanocomposite TENG. Reproduced with permission¹¹⁹. Copyright 2017, American Institute of Physics. **b** Mechanism of the starch paper-based TENG. Reproduced with permission. Copyright 2018, Springer. **c** Mechanism of TENGs based on CaCl_2 modified starch film. Reproduced with permission¹²². Copyright 2019, Elsevier. **d** Schematic representation of structure and the working principle of TENGs from thermoplastic starch-PDMS and the corresponding outputs for **e** human walking detection and **f** gait analysis. Reproduced with permission⁴⁰. Copyright 2011, American Chemical Society.

biodegradability, and crucial biological functions^{136–138}. Niu et al.¹³⁹ reported a gelatin/ poly (acrylic acid N-hydrosuccinimide ester) (PAA NHS ester)-based ionic conductive organohydrogel by introducing a glycerol water binary solvent system, which was used as wearable health monitoring device. The starch was also widely used in the preparation of gels can readily generate a three-dimensional network *via* physical entanglement of amylose and/or amylopectin¹⁴⁰. A large amounts of water or biological

fluids can be retained due to their good hydrophilicity. Furthermore, the starch chains may be easily cross-linked through the abundant hydroxyl groups. So, the starch-based gels were intensively investigated and shown great potential especially in biomedicine^{62,141–146}. Gonzalez et al.¹⁴⁷ prepared starch-based hydrogels through the Diels-Alder cross-linking reaction between furan modified starch and water-soluble bismaleimide. The photograph and SEM images were shown in Fig. 8a. Graphene

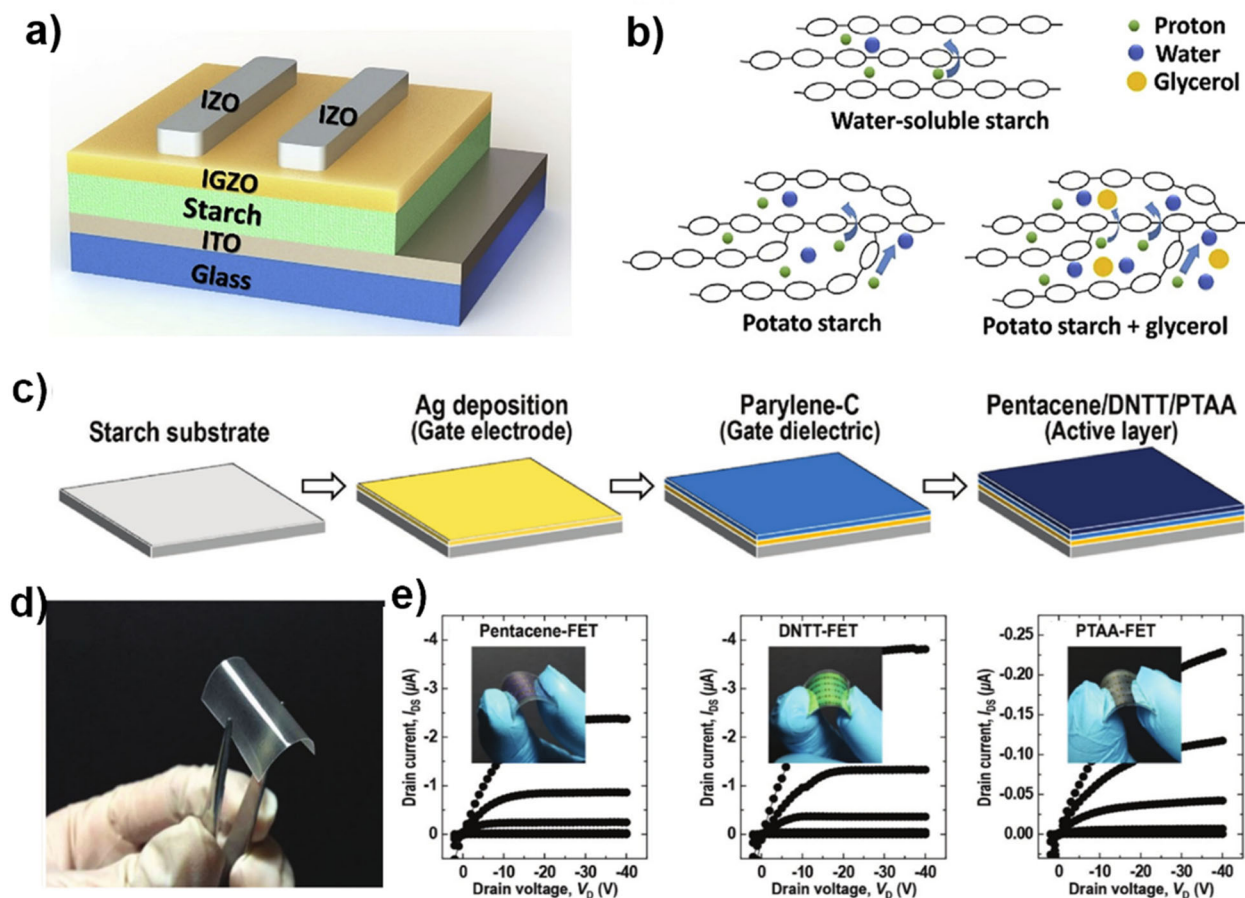


Fig. 6 Organic transistor based on starch films. **a** Device structure of the thin film transistors with starch as the ion-based gate dielectric. **b** Schematic illustration of the proposed ion (proton) transportation in the starch-based gate dielectrics. Reproduced with permission¹²⁸. Copyright 2017, Elsevier. **c** Schematic diagrams of fabrication process of the OFET based on starch-PVA film. **d** Photograph of the flexible starch-PVA film. **e** Output characteristics of various flexible devices fabricated on the starch-PVA film. The inset images in (e) show the fabricated devices and their flexible features. Reproduced with permission⁹². Copyright 2018, Wiley.

nanoparticles were introduced to endow the antibacterial and electrical conducting property to the hydrogel, which might find prospects in biomedical application. Recently, Lin et al.¹⁴⁶ prepared an adhesive hydrogel from ionically crosslinked starch, which had good electrical conductivity, high tissue adhesion, strong antibacterial ability. The wound condition diagnosis and management might be realized due to the conductivity of the hydrogel considering the wound condition can be transferred into electrical outputs. In addition, the application of starch-based gels in smart window was also explored¹⁴⁸. Wang et al. prepared smart window using thermosensitive starch gel as active layer, which shown an excellent solar modulation property of 17.9% and average light transmittance of 57.8%. The details of the reported FEs based on starch gels were listed in Table 2.

Sensing elements based on starch gels

Various sensors were fabricated on starch gels to obtain biocompatible devices^{149,150}. Kanaan et al.⁴⁵ synthesized a biodegradable semi-interpenetrating polymer networks (S-IPNs) based on starch and copolymer of 2-hydroxyethyl methacrylate (HEMA) and 1-butyl-3-metaprazolium chloride (BVIImCl) cationic (Fig. 8b). According to the relative composition of different monomers, the electronic and ionic conductivity of the hydrogel was manipulated in range from 0.1 to 5.2 S cm⁻¹, and the complex shear modulus was regulated in a range from 0.6 to 6.4 MPa. The flexible hydrogel worked as a multi-stimuli-responsive platform that was sensitive to changes of

relative humidity, ionic strength and current. Therefore, the adsorption/release capacity of the hydrogel toward *L*-tryptophan was regulated through the DC voltage applied. Furthermore, the hydrogel shown good biocompatibility due to the existence of starch. It may find widely applications in biological separation, sewage treatment systems, biomedical and electrochemical equipment. Liu et al.¹⁵¹ developed a flexible, ultra-low-cost conductive hybrid elastomer by mixing polydimethylsiloxane (PDMS) and starch hydrogel, which had strong elasticity (56 kPa) and acceptable conductivity (10⁻² S m⁻¹). The composite elastomer can detect stress (gauge factor 0.71), strain (gauge factor 2.22) and humidity (sensitivity of 1.2 × 10⁻⁶ S m⁻¹ RH %⁻¹). Moreover, the sensor can be used for monitoring finger movements and bottle grasping (Fig. 8c, d), demonstrating its potential in artificial skin and wearable devices. Noticeably, the volume ratio of the starch and PDMS in the composite was about 3:1, which is also significant for the cost reduction. Xu et al.¹⁵² selected potato starch as the main network and introduced polyvinyl alcohol (PVA) and borax to improve the mechanical and conductive properties of hydrogels. Due to the dual reversible interactions of hydrogen bonding and the boronic ester linkages, the hydrogels shown enhanced mechanical performance and super-fast self-healing ability. The hydrogel was used as a strain sensor with GF = 1.02 at 110–200% strains, which can quickly perceive human movements with a response time of 180 ms, even small movements such as swallows and sounds.

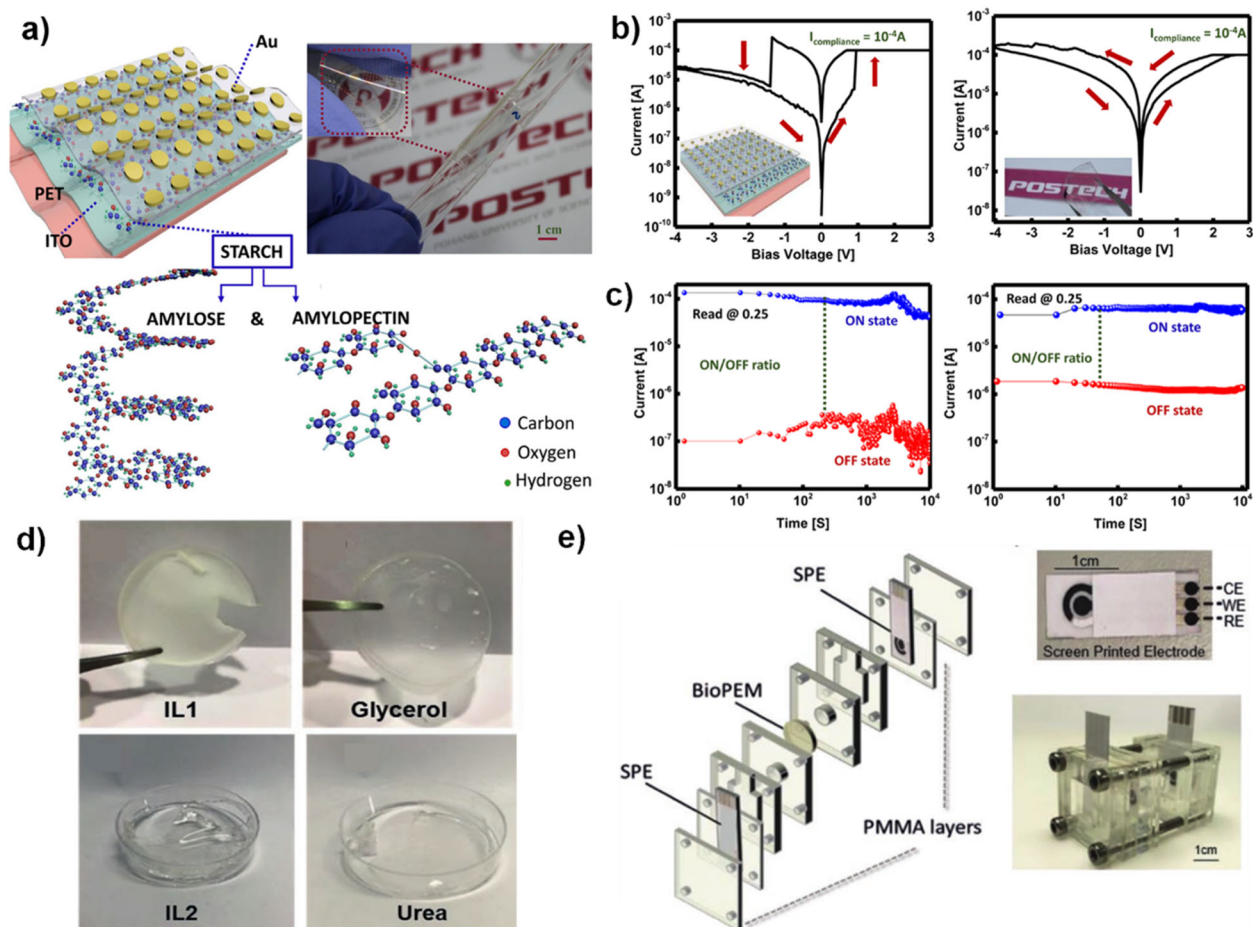


Fig. 7 Memory devices and batteries based on starch films. **a** Schematic illustration of the structure and the photograph of flexible ReRAM memory device on starch-chitosan. **b** I–V curves and **c** on/off ratio of the ReRAM devices composed of Au/starch/ITO/PET and Au/starch–chitosan/ITO/PET. Reproduced with permission³⁵. Copyright 2016, American Chemical Society. **d** Photographs of the starch based BioPEMs and **e** the corresponding battery cell. Reproduced with permission¹³⁵. Copyright 2020, Wiley.

The amylose and amylopectin may exhibit different mechanical property due to their structure difference. The linear chain of amylose makes it more resistant to processing and may have better mechanical performance and less water sensitivity¹⁵⁰. Liu et al.¹⁵³ prepared flexible, transparent, ionically conductive hydrogel by simply mixing high-amylose starch with CaCl_2 solution. This material shown adjustable mechanical strength (500–1300 kPa), elongation at break (15–32%), Young's modulus (4–9 MPa), toughness (0.05–0.26 MJ/m³) and suitable resistivity (3.7–9.2 $\Omega\cdot\text{m}$). The hydrogel can be used as a sensor to detect strain, moisture and liquid with different pH value. The Amylopectin with dendritic structure may supply more physical interaction with other materials thus is suitable to fabricate adhesive hydrogel. Gao et al.¹⁵⁴ combined amylopectin with poly (acrylamide–acrylic acid) (P(AAm–AAc)) to prepare a hydrogel with good adhesion, toughness and electrical conductivity. As a wearable sensor, the hydrogel shown high strain sensitivity ($\text{GF} = 6.93$) and stability, which was used to monitor various motions of human, such as joint bending, walking, jumping, even talking.

Supercapacitor based on starch gels

Due to the carbon-rich nature and low price, starch has been used as a common precursor for producing activated carbons which have wide applications such as electrode material^{155–157}. Wang et al.³⁴ reported a simple, rapid and scalable method for the production of the three-dimensional flexible and porous graphene

electrode materials, which simply dissolved starch in GO suspension followed by thermal carbonization and chemical activation. The electrode had a high specific surface area of $1,519 \text{ m}^2 \text{ g}^{-1}$, high energy density of 19.8 Wh kg^{-1} at the power density of 0.5 kW kg^{-1} and a high power density of 9.9 kW kg^{-1} at the energy density of 9.6 Wh kg^{-1} . The structural diagram and energy storage performance of the supercapacitor are shown in Fig. 9a. Furthermore, the flexible supercapacitors kept specific capacitance retention rate of 80% after 8000 cycles at 10 A g^{-1} . The supercapacitor worked well even under large angle bending (138°), thus may find potential in the application of wearable equipment. Similarly, Liu et al.¹⁵⁸ prepared three-dimensional interconnected reticular porous carbon electrode using corn starch as the carbon precursor (see protocol in Fig. 9b). The 3D electrode shown high specific capacitance of 372 F g^{-1} at a current density of 0.5 A g^{-1} in 2 M KOH electrolyte with high energy density and long cycle life. The cyclic C–V diagram is shown in Fig. 9c. Considering the high purity and low cost of corn starch and the excellent capacitance performance of 3D electrode, this study opens a way for the large-scale production of low-cost and high purity porous carbon for supercapacitors. Willfahrt et al.¹⁵⁹ prepared a screen printable hydrogel as electrolyte using corn starch and citric acid with ionic conductivity up to $2.30 \pm 0.07 \text{ mS cm}^{-1}$. The hydrogel shown excellent printability and prolonged stability against degradation. The specific capacitance of the printed supercapacitor was up to 54 F g^{-1} .

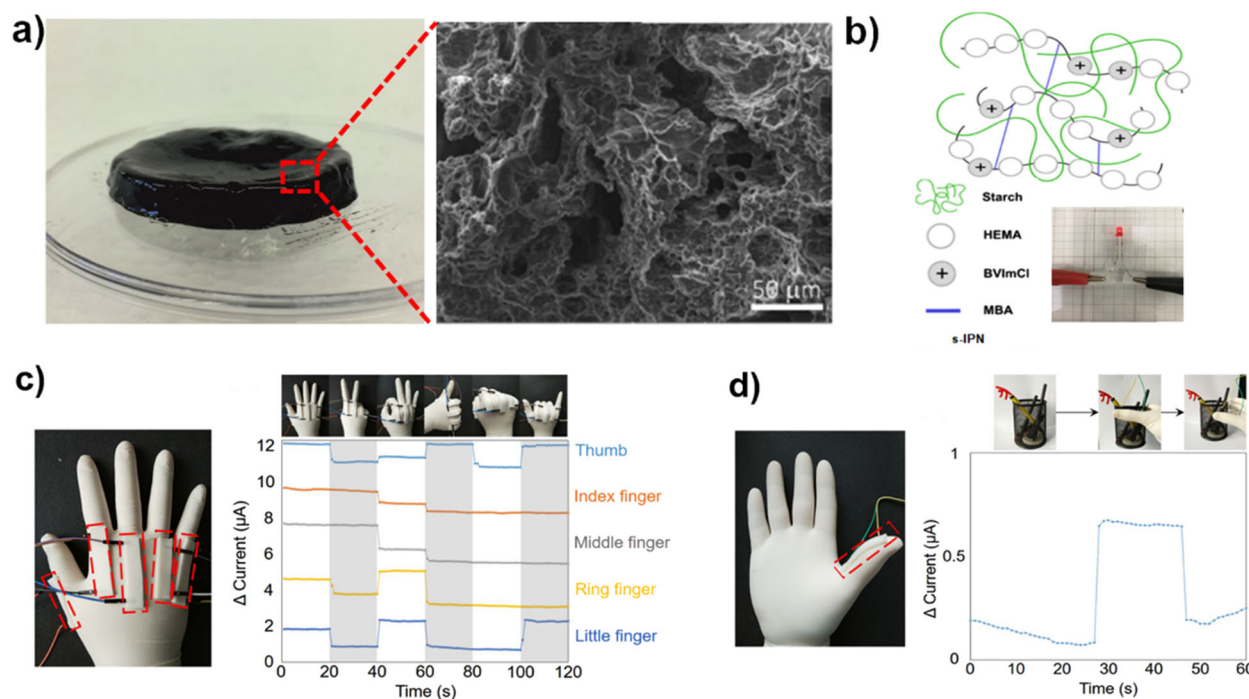


Fig. 8 Conducting and sensing elements based on starch gels. **a** Photograph and SEM images of the starch/graphene hydrogels. Reproduced with permission¹⁴⁷. Copyright 2018, Elsevier. **b** Chemical structure of the S-IPNs hydrogel with electronic and ionic conductivity. Reproduced with permission⁴⁵. Copyright 2019, American Chemical Society. **c** Photographs of starch hydrogel sensors on the glove and the responding current signal under different hand configurations; **d** photographs and signal of starch hydrogel sensor to monitor the hand grasping a pencil vase. Reproduced with permission¹⁵¹. Copyright 2019, Elsevier.

Table 2. Summary of FEs based on starch gels.

Device Type	Materials	Performance	Function of starch	Ref
Sensor/biomolecules carrier	Starch, poly (HEMA-co-BVImCl)	Electric responsive 2–5 V	Increase biocompatibility	45
Adhesive hydrogel	Starch, cross linkers	Viscoelastic antibacterial conductivity	Adhesives	146
Smart window systems	Starch, AgNWs, cellulose	Solar modulation 17.9%	Active layer	148
Sensor	Starch, PDMS	Stress (GF = 0.71), strain (GF = 2.22), humidity ($1.2 \times 10^{-6} \text{ S m}^{-1} \text{ RH}\%^{-1}$)	Substrate	151
Sensor	Starch, PVA, borax	Strain (GF = 1.02), $S = 180 \text{ ms}$	Network structure	152
Sensor	Starch, CaCl_2	Strain, moisture, pH	Network structure	153
Sensor	Amy, P(AAm-AAc)	Strain (GF = 6.93)	Assistant	154
Supercapacitor	Starch, GO	Energy density 19.8 Wh kg^{-1}	Carbon precursor	34
Supercapacitor	Starch, KOH	Energy density 24.5 Wh kg^{-1}	Carbon precursor	158
Supercapacitor	Starch, citric acid	Specific capacitance 54 F g^{-1}	Electrolyte	159
Supercapacitor/absorbent	Starch, GO	Specific capacitance 316 F g^{-1}	Modification of GO	41
Triboelectric nanogenerator	Starch, PDMS, silicone rubber	Open-circuit voltage $\sim 25 \text{ V}$	Electrode	39

Starch-based nanoparticle was also used in the preparation of supercapacitors. For example, starch nanocrystals prepared by hydrolysis of starch have been widely used in many fields^{62,63,65,160}. Chen et al.⁴¹ used graphene oxide (GO) nanosheets to combine with dialdehyde starch nanocrystals to fabricate porous, strong, compressible aerogels that were used as supercapacitor electrodes and highly efficient adsorbents (Fig. 9d). The starch nanocrystal prevented self-stacking between graphene nanopores to improve the electrical properties of aerogels, resulting in the increment of the specific capacitance from 198 to 316 F g^{-1} . The good mechanical properties and high specific surface area of the aerogels promised their application in dyes removing. Therefore, the hybrid aerogel might be used as a

flexible electrode of supercapacitors or candidate materials for biomedical and environmental cleaning.

Nanogenerator based on starch gels

A common limitation of the sensing system is that the most sensors cannot work without external power source. TENGs can act as an active sensor to detect pressure without the use of an external power source^{9,110,111}. In addition, complex circuits can also be avoided in these self-powered devices. Chen et al.³⁹ proposed a self-powered, flexible, triboelectric sensor (SFTS) composed of starch-based hydrogel, polydimethylsiloxane (PDMS) and silicone rubber. The two-dimensional SFTS (2D-SFTS) with a

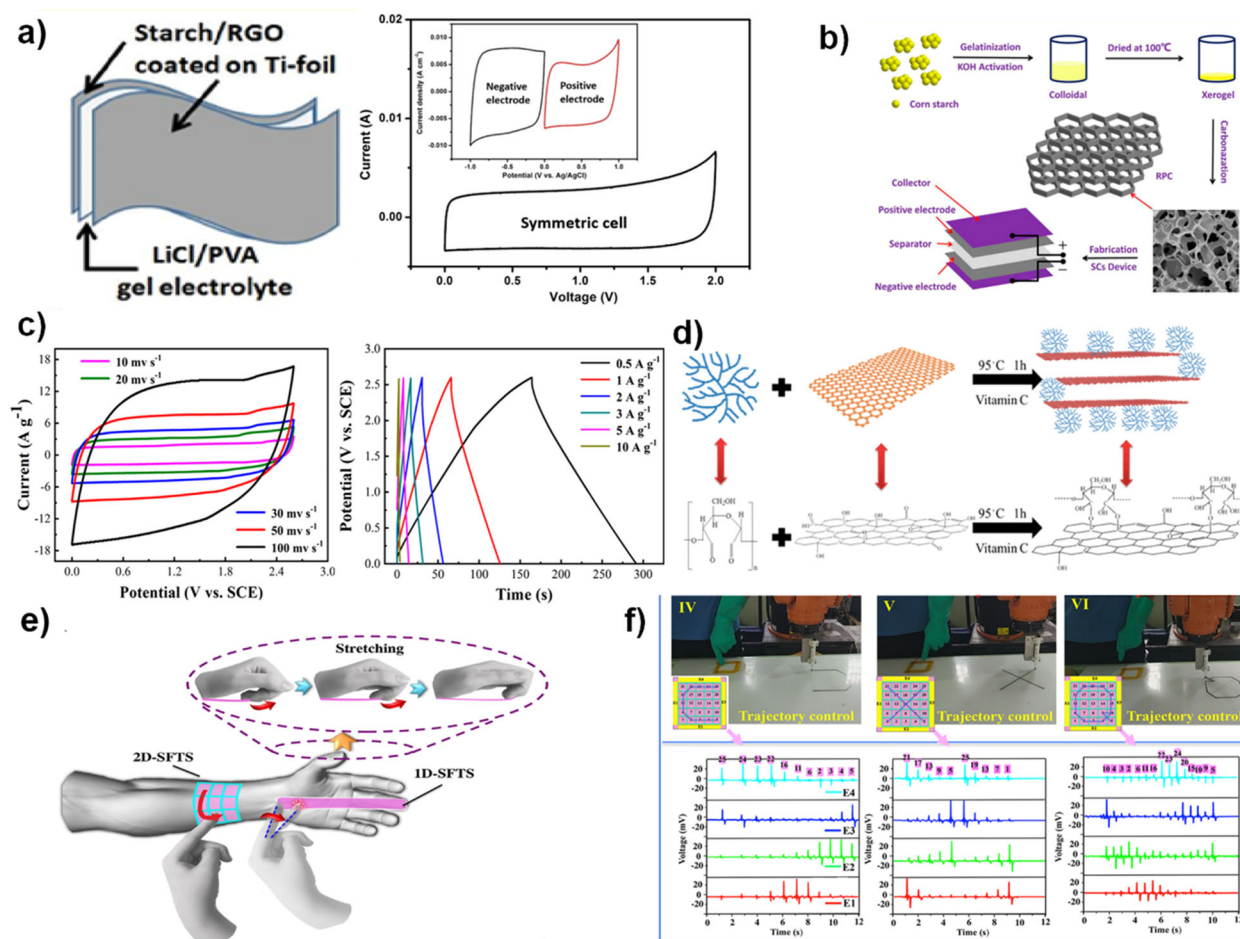


Fig. 9 Supercapacitor and nano generator based on starch gels. **a** Schematic diagram of supercapacitor and capacitive performances of starch/RGO and starch supercapacitor. Reproduced with permission³⁴. Copyright 2017, American Chemical Society. **b** Schematic diagram and **c** electrochemical performances of the supercapacitor with a 3D printed electrode. Reproduced with permission¹⁵⁸. Copyright 2019, American Chemical Society. **d** Aerogel made of GO and starch nanocrystal that can be used as supercapacitor electrodes and highly efficient adsorbents. Reproduced with permission⁴¹. Copyright 2019, American Chemical Society. Self-powered, flexible, triboelectric sensor (SFTS) for 3D motion detection and **f** 3D robotic manipulations. Reproduced with permission³⁹. Copyright 2018, American Chemical Society.

grid structure can track continuous sliding information on the fingertip, such as trajectory, velocity, and acceleration. 3D motion detection (Figs. 9e) and 3D robotic manipulation (Fig. 9f) were also realized by combining with a one-dimensional (1D) SFTS. By introducing starch, the cost was decreased and environmental friendliness was improved for the composite elastomer.

DEGRADABILITY AND BIOCOMPATIBILITY OF STARCH-BASED ELECTRONICS

The natural polymers including cellulose, lignin and starch belong to biodegradable polymer materials, which can be degraded by bacteria, mold, algae, and other microorganisms through enzyme or chemical hydrolysis. The main molecular chains are firstly broken resulting into the reduction of molecular weight. They eventually decomposed to small molecules or metabolized into carbon dioxide and water¹⁶¹. The polymers with higher molecular weight may takes a longer time to be degraded. A greater microbial diversity may also accelerate the degradation, such as the activated sludge.

The natural polymers such as starches usually have to be blended with synthetic polymers to reach the mechanical requirements in practical application. The degradation of the composite of starch and synthetic polymers may be accelerated because the starch might be firstly decomposed by

microorganisms, which makes the hybrids porous and increases the specific surface area, thus facilitating further enzymatic hydrolysis or decomposition in the nature¹⁶².

As known the starch is derived from nature and should be degradable, lots of experimental protocols have been established to confirm the degradation of FEs based no starch. One intuitive way is recording the decomposing process of the device. For instance, the conductive starch film developed by Miao and co-workers³⁸ was found to be quickly dissolved and disappeared in the buffer of acetic acid and sodium acetate (pH = 4.5) with 3wt % lysozyme after about 8 min, as shown in Fig. 10a. In another examples, the starch device was gradually covered with fungus until it was completely degraded when it was soaked in fish tank water (Fig. 10b)³³. In our previous work, the starch-based sensor was broken in the flush water generated by the shower nozzle and then completely decomposed in about 10 min, indicating a rapid degradation under water flushing (Fig. 10c)³⁷. Besides, the starch-based devices were also demonstrated to be degradable in static water^{37,120}. It should be pointed out that the treatment of the other materials in these devices were usually ignored, including the graphene, conducting polymers, or metals. The reason may be ascribed to their non-hazardous feature and low content of these conductive materials in FEs. However, it may be a risk when the usage of the starch-based FEs is large enough. The non-degradable components may be recycled by filtration or flotation

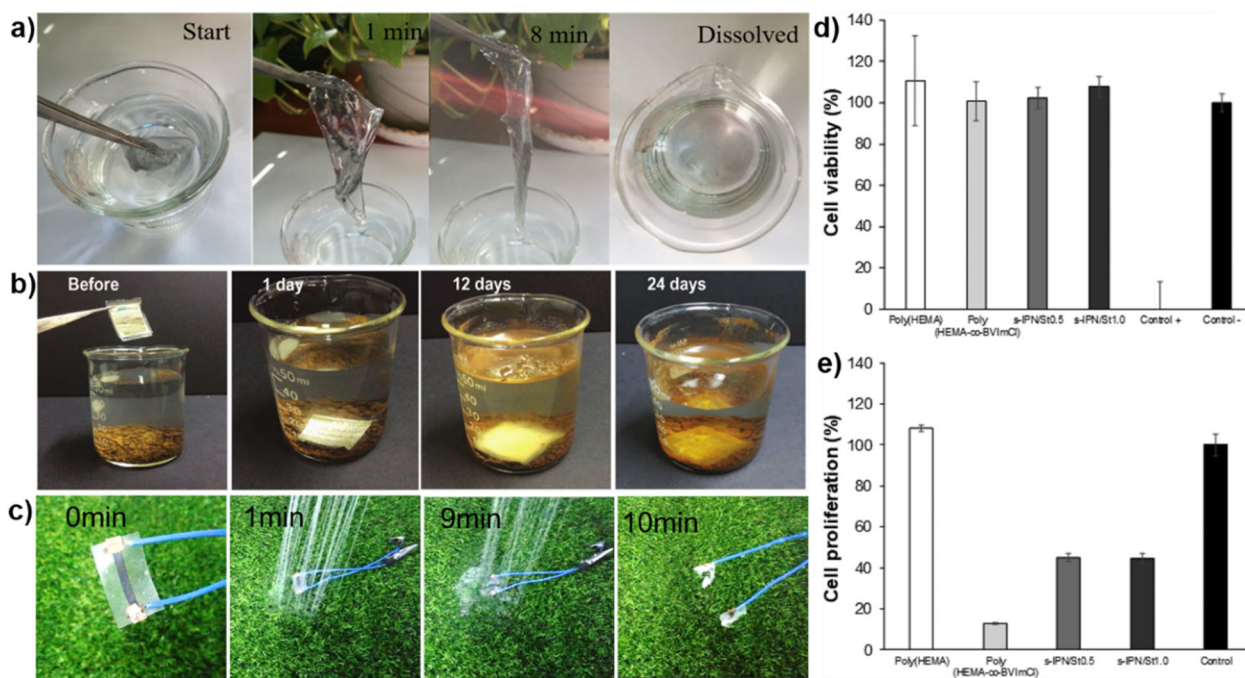


Fig. 10 Degradability and biocompatibility of starch-based FEs. **a** Dissolution behaviors of the starch-based conductive film. Reproduced with permission³⁸. Copyright 2018, American Chemical Society. **b** Biodegradability tests of the starch-based device in fishbowl water. Reproduced with permission³³. Copyright 2017, Wiley. **c** Degradation of the starch-based sensor under water flushing. Reproduced with permission³⁷. Copyright 2020, American Chemical Society. **d, e** The cell viability and the cell proliferation data of the hydrogels tested against fibroblasts. Reproduced with permission⁴⁵. Copyright 2019, American Chemical Society.

after the degradation of starch. But solid protocols need to be addressed in the future research.

Except for degradability, the biocompatibility of the starch-based FEs was also concerned especially for medical applications, which can be confirmed through cytotoxicity tests^{45,163}. The cell viability and the cell proliferation assays of the starch-based hydrogels were checked using Balb/3T3 fibroblasts cell in Kanaan's work (Fig. 10d, e)⁴³. The cell viabilities (~100%) for all starch-containing hydrogels after 48 h direct contacting were around 100%. The cell proliferation increased approximately 20% for the sample in the presence of starch. These results suggest that the FEs based on starch are degradable in natural environments and biocompatible in human body, thus may be free of e-waste and find applications in biomedical therapies.

CONCLUSIONS AND OUTLOOKS

In summary, comparing with traditional flexible materials such as PET or PI films, starch has significant advantages including low cost, controllable biodegradability, easy processability and acceptable biocompatibility, which make it versatile in future green electronics. Starch can work as flexible substrates, electrodes, carbon source, dispersants, stabilizers, or modifier of conductive materials. In the most of its applications, starch appears in the form of thin films or gels. The electronic products may be included but not limited as flexible electrodes, sensors, supercapacitors and nanogenerators. However, there are still some problems need to be resolved despite abundant recent breakthroughs have been made.

Firstly, the mechanical property of the starch films/gels need to be strengthened in some occasions when the mechanical robustness is required. Some physic and chemical strategies were developed to date. For an example, small molecules such as ethylene glycol was usually used as plasticizers of the starch film to improve the toughness³⁷. PVA with cross-linkers was added into the starch film to increase the tensile strength and solvent

resistance of starch substrate for FEs³³. In addition, the molecular chains of the starch can be grafted with polymers, which may also achieve high mechanical strength. But the chemical modification may also bring other problem, such as the degradability. The synthesized degradable polymers such as polylactic acid (PLA) or poly (butyleneadipate-co-terephthalate) (PBAT) may be good choices to reinforce the starch-based materials^{164–166}.

Secondly, the additional components in starch-based electronics may increase the concerns for their biocompatibility and hinder their biomedical applications. For instance, the metals, nanowires, or conductive polymers that were used to improve the electrical conductivity. The investigations on the toxicology of such materials in FEs need to be promoted urgently, in which the biologist should be involved. Some metals and metal alloys have been found to be bioabsorbable and widely used in temporary biomedical implants¹⁶⁷. On the other hand, if the conductive materials were chemical modified or enveloped with biocompatible molecules, the toxicity may be significantly lowered down to the acceptable level, thus meeting the requirements of diagnose or therapy.

Thirdly, the starch is degradable in the nature environment, but the complete degradation of FEs may not be practical. One way to reach the totally degradation may be the using of organic conductive polymers. Otherwise, the non-degradable materials including metals, graphene, carbon nanotubes need to be recycled in the future when the starch-base FEs are widely used. Zhou and coworkers¹⁶⁸ recycled liquid metal form FEs after dissolving the soft substrate, which supplied a clue for the recycle of the non-degradable component in the FEs.

Fourthly, starch-based electronics can be gradually degraded in water or biological fluid, also implying that they might be impacted by external environment during working. In general, the life-time of the FEs may be not too long. How to make their reliable life-time be controlled remains an open question. Spiridon et al.¹⁶⁹ studied the degradation behavior of PVA/starch composite film, finding that the enzyme and nanoparticles

influenced the degradation rate. Further investigation is highly desired in the future to explore more methods to manipulate the degradation of starch-based FEs.

Last but not the least, the self-powered systems or devices that do not require external power sources, may provide the possibility for reliable outdoor electronics. Starch-based self-powered electronics can provide independent, continuous power to the FEs. For instance, Sarkar et al.^{24,26} prepared triboelectric nanogenerator based on thermoplastic starch^{39,40}. Such products are increasingly favored by more and more applications, but their development is still in the infancy stage. More portable and sensitive self-powered devices need to be further explored.

The work is far from done, however, it is convincing that more and more practical FE products based on starch will be developed in the future with further efforts putting into this exciting area, since the great potentials have been widely recognized. It is also of the great significance to the global green and sustainable development that is concerned by human beings.

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AUTHOR CONTRIBUTIONS

H. Xiang wrote original draft and collected the data. H. Liu. conceptualized, supervised, and edited the draft. Z. Li, T. Chen, and H. Zhou reviewed the manuscript and provided scientific comments and suggestions. W. Huang provided overall supervision of the work.

COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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