Recent advances in highly integrated energy conversion and storage system

Changxiang Shao¹ | Yang Zhao² | Liangti Qu³

¹School of Chemistry and Pharmaceutical Engineering, Shandong First Medical University & Shandong Academy of Medical Sciences, Taian, Shandong, P.-R. China
²Key Laboratory of Photoelectronic/Electrophotonic Conversion Materials, Key Laboratory of Cluster Science, Ministry of Education of China, School of Chemistry and Chemical Engineering, Beijing Institute of Technology, Beijing, P. R. China
³Key Lab of Organic Optoelectronics and Molecular Engineering of Ministry of Education, Department of Chemistry, Tsinghua University, Beijing, P. R. China

Abstract
The vigorous development in the field of energy conversion and storage devices directly contributes to the full utilization and convenient use of clean energy. However, some drawbacks of independent energy conversion and storage devices, including unstable, insufficient energy output and dependence on external power supply, are difficult to overcome by self-optimization, thus, hindering their further development and direct application. Coincidentally, the combination of above two devices can solve these problems, which conforms to their intrinsic needs for development. At the same time, the pursuit of portability and miniaturization also promotes the development of the power system toward a highly integrated direction. Therefore, we introduce several integration modes of energy conversion and storage systems, with emphasis on all-in-one power system, possessing the highest integration in this review. From the aspect of device configuration, working mechanisms and their performances, the all-in-one power systems based on different energy sources (e.g., mechanical, solar, thermal, and chemical energy) are discussed and analyzed. Finally, the design strategies are summarized and the potential development directions in the future are proposed. This review aims to provide a comprehensive overview of highly integrated energy conversion and storage system, and seeks to point out the opportunities and orientations of future research in this field.

KEYWORDS
all-in-one, energy conversion, energy storage, high integration, power system

This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.
© 2022 The Authors. SusMat published by Sichuan University and John Wiley & Sons Australia, Ltd.
1 | INTRODUCTION

The contradiction between the increasingly serious global problems, including environmental pollution, energy crisis, climate change, and the urgent demand of human society to improve living standards, is becoming progressively prominent. In this context, the field of clean energy utilization has gained extensive attention. The related energy conversion and storage devices have also been widely concerned and developed rapidly in the last few decades.\(^1\)\(^-\)\(^4\)

The energy conversion device in a power system is responsible for collecting and converting the energy in the environment into easy-to-use electric energy. At present, a series of energy conversion devices have been explored. For example, solar cells,\(^5\)\(^-\)\(^7\) tribo/piezoelectric nanogenerators,\(^8\)\(^-\)\(^12\) thermoelectric devices,\(^13\)\(^-\)\(^17\) water-induced power generators,\(^18\)\(^-\)\(^26\) and other novel power producers have the ability to convert various energies, such as solar, mechanical, thermal, flow potential energy, into electricity based on different principles. In the process of energy conversion, the energy sources deriving from nature or human random activities have the characteristics of discontinuity, lability, and uncontrollability. As a result, the power generators based on these energy sources often produce small, unstable, and noncontinuous power output, which will seriously affect their power supply for electrical appliances.\(^27\) To alleviate these problems, people put forward methods such as enlarged preparation of devices. In addition, applying energy storage devices to store and reuse the electricity has become an important solution, which can not only improve the energy supply capacity, but also increase the stability of the power system.

Energy storage devices mainly, including supercapacitors and batteries, play the role of charge storage in power systems. The supercapacitors store energy by means of double electric layer or reversible Faradaic reactions at surface or near-surface electrode,\(^28\)\(^,\)\(^29\) while batteries usually store energy by dint of electrochemical reactions at internal electrode.\(^30\) These two types of energy storage devices have their own advantages and disadvantages in different aspects of power density, energy density, cycle stability, and service life, etc. Recently, their charge storage performances have been significantly improved through the device structural optimization and regulation of electrode, electrolyte, and separator materials, which has been widely reported.\(^31\)\(^-\)\(^33\) However, these energy storage devices cannot be charged by themselves, making them impossible to use at anytime and anywhere. Once the stored energy is consumed, the device needs to be charged by external power. To achieve self-charging, the method of combination with energy conversion devices is put forward.

As can be seen from the above introduction, both energy conversion devices and storage devices have their inherent defects that are difficult to be solved by simple self-optimization. The combination system of these two kinds of devices can make up for the defects of each other and make them offer better performance as power supply devices. Therefore, more attention has been paid to the integrated system of energy conversion and storage devices. With the development of portable and miniaturization of electronic devices, people have also put forward higher requirements on the integration degree of combination systems. The well-designed, highly integrated system in Figure 1 is expected to be designed and fabricated, which can clearly reduce supplemental electronics for the power management. The application significance of a highly integrated power system is mainly embodied in the realization of an intelligent self-power system. The self-powered system will play an important role in a variety of applications from microscale (wearable/implantable/portable electronics and micronano fluid devices) to macroscale (outdoor mobile charging systems, electric vehicle power supply, climate big data detection, and health monitoring systems), preparing for the era of Internet of Everything.

To deal with the demand, power systems with different integration degrees are investigated. As research continues, a lot of innovative work about highly integrated power systems has been reported. Therefore, we will briefly introduce the development of integrated energy conversion and storage systems and focus on power system with a high degree of integration, namely all-in-one power system. This review will present a critical review of the current and significant progress in all-in-one power devices based on different energy sources. Through the analysis and discussion of the materials selection, device configuration, work mechanism, and performance involved in all-in-one power system, the design ideas are summarized and the
future research directions are also proposed. This review is expected to provide references and ideas for the following research, which offers direct services for the construction of a new integrated energy conversion and storage system.

2 | THE DEVELOPMENT OF INTEGRATED POWER SYSTEM

As mentioned earlier, since energy conversion and storage devices work on different physical or chemical mechanisms, it is difficult to integrate two completely different units. Because, the research of a highly integrated all-in-one power system is not accomplished overnight, but after a long period of active exploration and hard research.

2.1 | The integrated power system with independent devices is connected by external wires

Initially, the simplest and easiest method to combine the energy conversion and storage devices is to connect two separate device units via external circuitry, which allows the converted energy to be stored and available on demand. The previous reports usually place emphasis on the preparation of single energy conversion or storage devices, and then combine them with commercial energy storage or conversion device if needed.34–37 As shown in Figure 2A, laser-assisted microsupercapacitors array can be charged by commercial solar cell and then power LED.36 In addition, the paper-based microsupercapacitors (Figure 2B) fabricated via templating and imprinting strategy can store the energy generated by wind turbines.37

Since the combination of homemade and commercial devices is prone to incompatibility, researchers carry out studies of power generation and storage at the same time.38,39 For example, the tribo/piezoelectric nanogenerator and electrochromic microsupercapacitor are made to produce an intelligent self-charging power pack, which can self-charge to 3 V under external mechanical motion.38 A series of similar researches about integrated devices that adapt to different application environments are explored, making a great contribution to the development of power devices.

In a word, this combination mode relies on independent energy conversion or storage devices and is easy to design and has great autonomy and controllability in the selection and assembly of devices. However, the problems are still unavoidable such as low integration, large space occupation, low overall energy storage efficiency, and high energy management costs.

2.2 | The in-plane integrated power system

Later, with the rapid development of wearable and portable electronics, the planer macro-/microenergy devices have attracted widespread attention. The construction of
in-plane energy devices with interdigital or helical electrodes structures is achieved by means of various technologies, such as laser micrano processing, template imprint, screen printing, photolithography, three-dimensional (3D) printing, and so on. In recent years, our group prepared a variety of planar supercapacitors and batteries mainly based on laser direct writing or laser-assisted technology. These diverse devices have been applied in different scenarios, including aqueous antifreezing and heat-tolerant microsupercapacitor, capacity recoverable zinc-ion microsupercapacitor, swallowable and ingestible microsupercapacitor, the first flexible dual-ion microbattery with superior capacity and ultrahigh energy, aqueous Zn-MnO2 rechargeable microbattery. Moreover, the microsupercapacitor and wireless coils are seamlessly integrated in Figure 2C. The integrated system realizes moveable charging and lays a foundation for contactless microelectronics and flexible microrobotics. As for energy conversion devices, based on the phenomenon of moisture-induced electricity generation first reported by our group, we further prepared planar generators through structural optimization. For example, the graphene-based hygroelectric generators with rollable and stretchable features in Figure 2D are fabricated by direct laser writing process, which is expected to provide a new pathway for the development of flexible moisture-electric converters and applications in intelligent platform.

As can be seen from the reported researches, the rapid development of single in-plane microenergy device by advanced manufacturing processes naturally accelerates the integration of different device units in a single plane. The in-plane integrated power system means that different device units are fabricated on the same plane. A typical in-plane energy system composed of photocatalytic fuel cells and asymmetric microsupercapacitors is exhibited in Figure 2E. The maximum power density of the fuel cell produced by 1 M urea solution is 3.04 μW/cm² under an ultraviolet intensity of 30 mW/cm². The area capacitance of asymmetric supercapacitors is as high as 54.7 mF/cm² at the current density of 0.5 mA/cm². Four fuel cells and two supercapacitors can be easily constructed into an energy chip to directly power small electronics. This eco-friendly, self-contained system has great potential for future portable electronics applications.

In short, this integration mode is more in line with the development trend of miniaturization of electronic devices, and is easy to integrate multifunctional electronic components. However, the system preparation often relies on advanced high-precision processing instruments. Thus, the great leap forward in energy integration systems is hard to escape from the limitations of advanced manufacturing processes.

2.3 | The integrated power system with common electrode

The above integrated power systems are just different in the connection type of the two kinds of devices, and do not integrate the energy conversion and storage functions into a single device. To solve this problem, the researchers present a compromise between independent devices connected by external wires and a single device with synchronous energy conversion and storage: the two devices sharing one electrode. At present, it has become a widely recognized and effective strategy to combine different devices, which can not only improve the space utilization efficiency, but also increase the volume energy density of the whole system.

The integration forms with common electrode are varied in different energy systems. In one case, the devices are usually configured in a back-to-back three-electrode mode, in which the common electrode is actually a double-side electrode acting as a bridge connecting the separated energy conversion and storage system. Besides, a good example of another common device configuration is shown in Figure 2F, which is a system covering a moist-induced generator and a supercapacitor. They are integrated by sharing a single electrode to solve the problem of pulse electrical signal and small current output of the individual moist-induced generator. In addition, another situation occurs in fibrous energy devices which are usually constructed with coaxial structure. For example, vertically aligned TiO2 nanotubes covered with Ti wire serve as the common electrode of the solar cell and the supercapacitor. The two parts are coated with corresponding electrolytes and wrapped with counter electrodes, respectively. As depicted in Figure 2G, the solar cell generates electricity under sun irradiation, which charges the supercapacitor through other two electrodes connection. When the supercapacitor is fully charged, the two electrodes are disconnected. At this point, the supercapacitor can power external devices even under dark conditions. Unfortunately, the overall solar energy conversion and storage efficiency reaches 0.82%, suggesting there is much room for improvement. Obviously, integrated devices still face great challenges in how to better coordinate the performance of the two components to achieve higher energy conversion and storage efficiency.

Through the introduction of the above three combination modes of power systems, we can draw a preliminary conclusion that the way of independent devices connected by external wires (Figure 3A) is difficult to meet the development trend of miniaturization and integration of electronic devices. Benefitting from their own advantages, the in-plane integrated system (Figure 3B) and integrated energy system with common electrode (Figure 3C)
become the mainstream at present. Although these studies have greatly promoted the integration of energy devices, they are far from reaching the highly integrated system shown in Figure 3D. In recent years, with the deepening understanding of mechanisms and the continuous exploration of device design, the completely all-in-one power systems capable of electricity generation and storage have emerged. The following sections will focus on such systems and highlight the latest developments based on different energy transformations.

3 | ALL-IN-ONE POWER SYSTEM

3.1 | All-in-one power system based on mechanical energy

In recent decades, breakthrough progress has been made in piezoelectric-driven all-in-one power system for energy conversion and storage. Prof. Zhonglin Wang and his team have made great contributions to promote the development of this field. In 2012, they first proposed a new mechanism to simplify the two mechanical-to-electrical and electrical-to-electrochemical processes into one mechanical-to-electrochemical process, realizing synchronous energy conversion and storage. Based on the new design strategy, they fabricated a self-charging Li-ion battery as shown in Figure 4A.62 The key point of device design is to replace the common polyethylene separator in Li-ion battery with the piezoelectric material, such as piezoelectric polyvinylidene fluoride (PVDF) film. The well-designed device utilizes the piezo-potential generated by PVDF to drive the electrochemical reaction for achieving the self-charging effect under deformation. The device voltage is charged from 327 to 395 mV in 240 s at a pressure frequency of 2.3 Hz, and discharged to 327 mV at a current density of 1 μA for 130 s. Thus, the stored energy is about 0.036 μAh. The presumed working mechanism is shown in Figure 4B. First, PVDF film generates potential difference with positive piezo-potential at the cathode side and negative piezo-potential at the anode side under the action of pressure. Driven by a potential difference, Li⁺ in the electrolyte migrates from cathode to anode for screening the potential difference. Due to the directional movement of Li⁺, the Li⁺ concentration increases in the anode and decreases in the cathode, respectively. The local changes in Li⁺ concentration will upset the original chemical equilibrium. To reach a new chemical equilibrium, the electrode reactions take place. At the cathode, Li⁺ will be released from LiCoO₂ through the deintercalation reaction of LiCoO₂ ↔ Li₁₋ₓCoO₂ + xLi⁺ + xe⁻, and the generated electrons will remain in the current collector. While the Li⁺ at the anode are consumed by reacting with TiO₂ via the reaction of TiO₂ + xLi⁺ + xe⁻ ↔ LiₓTiO₂, leaving positive charges at the current collector. To keep charge neutrality and the continuity of the charging reaction, extra free electrons shift from cathode to anode during this process. Since there is no connection between two electrodes in the external circuit, there is no possibility of electrons being transferred from the external circuit, so it is assumed that electrons are transferred inside the battery in the form of leakage current. When chemical equilibriums at the two electrodes have been established, the reaction will stop. This means that mechanical energy is converted directly into chemical energy and stored in the battery. Based on the above principle, a series of self-charging Li-ion batteries have been constructed by regulating the electrode material, piezoelectric material, or electrolyte.63–66 Recently, it has been found that this design concept can also be applied to Na-ion battery. This hybrid device with soft property can
be charged to 0.65 V under mechanical bending in 150 s or palm patting within 300 s. It not only shows application potential in wearable devices, such as conformable skin sensors, smart healthcare devices, roll-up displays, and wearable communication units, but also proves the universality of the design strategy for integrating piezoelectric generators and batteries.

Compared with batteries, supercapacitors have higher power density with fast charging and discharging rates, which can meet some specific application scenarios. Therefore, Wang’s team introduced this strategy into the piezo-supercapacitor integrated system to develop self-charging supercapacitors in 2015. The all-in-one self-charging supercapacitor is designed in a similar way with replacing conventional separators with piezoelectric materials. As shown in Figure 4C, a completely integrated self-charging device is constructed with MnO$_2$ as the electrode of the supercapacitor, PVDF-ZnO as the separator and piezoelectric material, poly (vinyl alcohol)-H$_3$PO$_4$ as the gel electrolyte. The device can be charged to 110 mV in 120 s with compressive force and the calculated capacitance is about 0.2575 F g$^{-1}$. The self-charging process of the device in Figure 4D is similar to the above analysis. When compressive stress is applied, the PVDF film with piezoelectric effect creates a potential across the film. This potential difference drives the ions in the electrolyte toward the positive and negative electrodes, which will balance the generated piezopotential. The ions’ movement causes the electrochemical imbalance between the electrolyte and the positive and negative electrodes. To achieve a new chemical equilibrium, the redox reaction will occur on the surface of the positive and negative electrodes, thus, realizing the electrical energy storage process. After that, different
electrode materials (e.g., carbon nanotubes, graphene, carbon cloth, MoSe₂, NiCoO₂@ activated carbon cloth, NiCoOH-CuO@Cu foil, CoFe₂O₄@carbon cloth), electrolytes containing water electrolyte gel and ionic liquid electrolyte, piezoelectric materials including porous or electrospinning PVDF, BaTiO₃, K₀.₅Na₀.₅NbO₃, perforated swim bladder, are used to prepare self-charging supercapacitors with symmetric or asymmetric electrodes configuration.⁷⁹–⁸⁷ Notably, although these studies demonstrate the feasibility of self-charging behavior to some extent, further experimental data on energy conversion and storage mechanisms are still lacking, which limits the improvement of energy conversion efficiency. To better explore the mechanism of the self-charging process, Kim et al. directly detected the piezoelectric electrochemical effect in the self-charging process of supercapacitor by piezoelectrochemical spectroscopy. And the well-designed all-in-one supercapacitor can self-charge to a maximum of 207 mV. This research provides an important experimental basis for understanding the self-charging process of piezoelectric electrochemistry, and shows direct evidence for the role of piezoelectric electrochemical phenomena in the energy conversion and storage process.⁷⁸

In short, such an all-in-one device based on piezoelectric effect is achieved by replacing the separator in ordinary supercapacitor or battery with piezoelectric material (Figure 4E). However, this type of integrated device is still in the early stages of research with most of the work focused on proof-of-concept. The overall energy conversion efficiency is still low, and the saturation voltage of the energy storage unit is far less than the voltage provided by the piezoelectric generator. Therefore, further research should pay more attention to the mechanism investigation in the integrated system, so as to effectively guide the performance improvement of devices in efficiency stability and other aspects.

### 3.2 All-in-one power system based on thermal energy

There is a large amount of waste heat and low-grade heat in the surrounding environment. If this part of energy is utilized and converted into electricity efficiently, it will greatly promote the development of clean energy. Lots of heat-to-electricity technologies are based on the heat engine, while thermogalvanic and thermomagnetic effects have been vigorously developed. Among them, the Soret effect refers to the phenomenon that the thermal diffusion of components in homogeneous ionic solid or electrolyte under the action of temperature difference eventually results in concentration gradient and corresponding thermal voltage.⁷⁹,⁸⁰ Similar to the electronic Seebeck effect, the thermal voltage is determined by the temperature difference across the devices and the ionic Seebeck coefficient. Recently, Soret effect induced power generation and storage within a single device, which is regarded as thermally induced self-charging supercapacitor, aroused a great deal of interest.⁸¹

As shown in Figure 5A, the difference between the all-in-one thermally induced self-charging supercapacitor and ordinary supercapacitor lies in the electrolyte material. In addition to the function of ion transmission, the electrolyte material in all-in-one supercapacitor with Soret effect can produce a potential difference under the stimulation of temperature difference. The working mechanism of thermally self-charging supercapacitor is schematically shown in Figure 5B, which can be mainly divided into four processes. (i) First, the supercapacitor with uniformly distributed ions of electrolyte is in an uncharged state. Then the temperature difference is applied on both sides of the device, the temperature gradient between two electrodes drives more mobile positively charged ions to diffuse to the cold side, leaving less mobile negatively charged ions behind. Due to the quiet difference in thermodiffusion rate between the two kinds of ions, the concentration of different charged ions at the two electrodes is different, thus, forming a potential difference. (ii) Then, the two electrodes of the supercapacitor are connected by an external circuit while the temperature difference is ensured. The electron flow from the low potential electrode to the high potential electrode will be generated to balance thermoelectric potential. The charging speed is controllable through changes in external load resistance. This is the most important step for self-charging, and the transferred electrons are stored in the electrode via forming a double layer or initiating a pseudocapacitance reaction. The charging process is completed when the potential difference stored in the electrode is balanced with the generated thermoelectric potential. (iii) After charging, the temperature difference is no longer maintained and the connection of the two electrodes is disconnected. Due to the vanishing temperature gradient, the thermoelectric potential will disappear with the back migration of cations. At this point, the open-circuit voltage of the supercapacitor only depends on the charge stored in the electrode, which is considered as a charged state of supercapacitor. (iv) Finally, the supercapacitor is discharged by connecting electronics, the stored electrical energy is gradually consumed. Notably, the above processes, including the self-charging and discharge process of a typical Soret effect induced self-charging supercapacitor, are repeatable, demonstrating the strong potential of devices to derive power from intermittent heat sources.²²

Based on above principle, Zhao et al. first constructed an ionic thermoelectric self-charging supercapacitor in 2016, which realized the conversion of heat energy into...
electricity and storage within a single device. The supercapacitor applies carbon nanotubes modified Au as electrode and polyethylene oxide (PEO)-NaOH solution as electrolyte. The PEO-NaOH solution is composed of negatively charged polymeric chains and positively charged Na\(^+\) cations, which have extremely weak and strong mobility, respectively. The ion Seebeck coefficient of PEO-NaOH solution with noncompensated thermodiffusion of cations and anions is +11.1 MV/K. This innovative work analyzes the conversion efficiency of heat to electricity, which provides many ideas and has important reference value for the following research work. However, the disadvantages of liquid electrolyte, such as serious self-discharge, restrict its application in wearable fields. To deal with this problem, Kim et al. used solid-state polystyrene sulfonic acid film and polyaniline (PANI)-coated carbon materials to build a sandwich structured supercapacitor. The thermally driven ion diffusion with asynchronous ion diffusion rates induces voltage generation and following electrochemical reactions of PANI, thus, realizing self-charging behavior without external power supply. The supercapacitor can generate 38 mV with a large areal capacitance (1200 F/m\(^2\)) under the temperature difference of 5 K. Later, the thermally chargeable supercapacitors with similar structures were constructed using graphene oxide and polystyrene sulfonate acid as electrolyte, reduced graphene oxide and conjugated conducting polymer as electrode materials. The test results were similar to the processes mentioned above, indicating that this strategy is an effective method to construct an all-in-one self-charging power device.
In addition to the above-mentioned Soret effect induced all-in-one energy devices, the integrated energy conversion and storage system can also be realized through thermogalvanic effect. As shown in Figure 5D, the design principle of this kind of system is to choose appropriate redox species with similar redox potentials but opposite Seebeck coefficients, as the anolyte and catholyte. The cathode redox species with high potential become lower with increasing temperature due to their negative Seebeck coefficient, while anode redox species with low potential become higher with increasing temperature due to their positive Seebeck coefficient. In this process, temperature is the driving force to maintain the alternating potential difference of the power device, thus, realizing the simultaneous energy collection, storage, and transport. The energy output of an electrochemical cycle is the area of the loop in Figure 5D. According to the design idea in Figure 5E, \( \text{K}_3\text{Fe(CN)}_6/\text{K}_4\text{Fe(CN)}_6 \) and \( \text{KI}_3/\text{KI} \) with similar redox potentials and competitive Seebeck coefficients are selected as the electroactive species for solar-driven regenerative electrochemical cells. The device constructed from two redox pairs verified the above concept, and a potential differential of 62 mW can be achieved under a low temperature gradient of 35°C generated by sunlight.\(^{88}\) It is anticipated that the attempt with thermally driven ion electrochemical reactions initiates a new research direction in thermal energy harvesting.

In a word, the above all-in-one power system provides effective methods to achieve the transformation and storage of the low-quality heat energy into electricity. At present, the design ideas of integrated devices mainly include two types. One is to use appropriate materials with Soret effect as electrolyte for supercapacitor, and the other is to choose appropriate Redox species with similar redox potentials but opposite Seebeck coefficients as the anolyte and catholyte, respectively. The integrated systems with innovative design and concepts still need to study the working principles of each component. As a result, a systematic and deep understanding of the working mechanism is very important to improve efficiency. In addition, new materials and device configurations should be further optimized to meet the low cost and high-performance requirements of actual mass production.

### 3.3 All-in-one power system based on solar energy

Because of the intermittent nature of solar energy, energy storage is necessary in systems that are powered by collecting solar energy. The solar energy enabled integrated power system which has been widely studied in the last few decades. According to the different energy conversion modes in the charging process, the power system can be divided into photovoltaic charging system and photocatalytic charging system.\(^{89,90}\) The integrated photovoltaic charging system is usually achieved by sharing one electrode, since the charging and discharging processes are independent and different. The photo-induced redox reactions occur during energy storage process in photocatalytic charging system. In general, the charge generated by photo stimulation can be stored in a variety of ways, such as electric double layers at the electrode-electrolyte interface and redox reactions of electrode materials, fast and reversible reactions of redox couples in electrolyte, so this all-in-one system can be designed as a highly integrated two-electrode system.

One strategy for building the all-in-one power system is utilizing dual-function electrodes with photoelectric and capacitive storage properties to realize energy storage via forming electric double layers at the electrode-electrolyte interface or starting redox reactions of electrode materials. For example, activated carbon, as a part of electrode, is combined with dye adsorbed semiconductor layer and an intermediate layer of a hole-trapping compound to form the photoelectrode. The sandwich-like device is composed of a dual-functional photoelectrode and a counter-electrode made of activated carbon. In principle, the generated charges under the illumination are transferred to the activated carbon and eventually form double electric layers on the surface. The device can be charged to 0.45 V with a capacitance of 0.69 F/cm\(^2\).\(^{91}\) In addition, the devices with conducting polymer and porphyrin dye as the photosensitive anode electrode and porous activated carbon as counter electrode in Figure 6A have also been shown to store electrical energy converted from solar energy. The charging process of the device is explained as follows: the photo-excited charges concentrate on the conducting polymer and charge the anode under open circuit conditions. The ions in the electrolyte are oxidized by positive charge, and the oxidized ions diffuse to the counter electrode, thus, forming electric double layer on the surface of the activated carbon. Thus, the device can output electricity for a period of time even without light irradiation.\(^{92}\) In addition, metal oxides with pseudocapacitance properties can produce higher capacitance through reversible redox reactions.\(^{93-95}\) For instance, the TiO\(_2\) nanowire/NiO nanoflakes and the Si nanowire/Pt nanoparticle composites are served as photoanode and photocathode to prepare a solar-powered device in Figure 6B. It can be charged to \( \sim 0.45 \) V and yield a capacitance of \( \sim 455 \) F/g by pseudocapacitance of NiO without any external power supply. Moreover, the device becomes an integrated self-powered chemical sensor by adding the function of glucose detection, which provides a good reference for the construction of all-in-one energy conversion-storage-utility system.\(^{99}\)
What’s more, the semiconductor materials and their composite materials with both photoelectric and capacitance characteristics have been widely studied in the photoelectric capacitor. The utilization of these materials as dual-function photoelectric electrodes to construct all-in-one power devices is one of the important ways to realize light energy conversion and storage. Therefore, optimization and improvement have been made in the design of optical electrode from the aspects of microstructure control and material selection. For example, the material of two-dimensional (2D) cyanamide-functionalized polyheptazine imide is reported for the first time to achieve seamless combination of light capture, charge carrier separation and energy storage within one single material. The functions realization depends on the mechanisms of photoreduction of the carbon nitride backbone and alkali metal ions absorption-induced charge compensation. Therefore, the material can be used as the anode of the battery in Figure 6C to store photoinduced charge in the form of electrons for several hours. These interesting features can be applied to a wide range of optical driven devices, such as electrochromic devices, optical drive switches, and optical detectors. Besides, the 3D Cu foam@CuOx@NiCuOx nanosheets arrays combining photo-sensitive characteristics of CuOx and pseudocapacitance characteristics of NiOx are fabricated and show a significant photoenhanced pseudocapacitance effect. Compared with nonillumination condition, the areal specific capacity of 1.452 C/cm² under light illumination increases by 44.8%, which is attributed to the synergistic effect of a built-in electric field enhancement and electron holes migration enhancement mechanism. Moreover, the materials, including hexagonal-phase WO₃ and nanoporous Cu@Cu₂O hybrid array, also have been developed for the photoelectrodes investigation in three-electrode system. And the results show 38 and 37.9% capacitance improvement under light illumination. These researches provide theoretical basis and possibility for understanding and constructing new all-in-one light-assisted energy devices. And the photoenhanced capacitance characteristics of these supercapacitors also prove their potential applications in photo detectors.

In addition to supercapacitors, batteries can also be combined to store energy. Unlike supercapacitors, solar charged batteries rely on the redox reactions at the photoelectrode–catholyte/anolyte interface or at internal electrodes. The selection of appropriate redox couples in electrolyte plays an important role in device construction and performance improvement. For example, a lead-organohalide electrolyte CH₃NH₃I·PbCl₂ was explored to
prepare a dye-sensitized solar cell with in situ energy storage function. The coupling of lead and iodine in one electrolyte enables photoelectric-chemical reactions and chemical reactions to occur within a single device. The I\(^{-}/I_{3}^{-}\) redox couple is used to generate photovoltage under the illumination. And then, as shown in Figure 6D, the photopotential causes the oxidation and reduction reactions of Pb\(^{2+}\), thus, realizing the charging process. During the discharge process, reversible redox reactions will occur. The conceptual design for all-in-one power devices could provide an efficient way to utilize renewable solar energy.\(^{101}\) The redox reaction of internal electrodes is a more common way to realize the energy storage. The WO\(_3\) and Cu\(_2\)S nanomaterials were introduced into photoanode of dye-sensitized solar cell to facilitate de/lithiation of Li\(^+\).\(^{102,103}\) The device in Figure 6E adopting covalent organic framework integrating naphthalenediimide and triphenylamine units as cathode can couple photo-induced charge transfer and reversible de/lithiation electrode processes, implementing direct solar-to-electrochemical energy conversion and storage.\(^{104}\) Further test results demonstrate that different types of batteries, including Li-S battery,\(^{105}\) Li-I redox battery,\(^{106,107}\) can be coupled into all-in-one solar charging devices. From another perspective, solar energy can be utilized to optimize the oxygen evolution reaction kinetics. As shown in Figure 6F, BiVO\(_4\) or \(\alpha\)-Fe\(_2\)O\(_3\) photoelectrodes are used as air electrodes to improve the performance of the zinc-air battery, achieving record low charging voltage of 1.20 and 1.43 V, respectively. The holes created by the photoelectrode migrate to the surface and oxide water to oxygen during charging process. Due to the valence band potential of photoelectric electrode is higher than the \(O_2/OH^-\) couple potential, BiVO\(_4\) and \(\alpha\)-Fe\(_2\)O\(_3\) photoelectrodes with appropriate band structure are selected to promote oxygen evolution reaction, which opens up a new way for solar energy utilization.\(^{108}\) However, this study only realized the light-assisted charging and discharging of zinc-air batteries, and the entire photocharging process needs to be further explored.

In conclusion, the synchronous conversion and storage of solar energy can be realized by an all-in-one power system via dual-function electrodes construction or appropriate electrolyte selection to construct. Under the synergistic effect of energy storage materials and photoelectric materials, the device can be fully or partially charged via double electric layer at the electrode-electrolyte interface, redox reaction on the surficial/interior electrode or in the electrolyte. At present, the photocharging performance is still at a low level and the overall efficiency is rarely discussed. There is still a long way to go in optimizing device unit performance and device structure.

### 3.4 All-in-one power system based on chemical energy

The fuel cell, an energy conversion device that converts chemical energy into electric energy, has attracted extensive attention due to its high energy conversion efficiency and pollution-free characteristics. However, the fuel cell is still limited by its low power density due to the slow catalytic reaction kinetics at the cathode and anode. With the rapid development of electrical equipment, such as electric vehicles in recent years, there is an urgent demand for power devices with both high energy density and power density. In addition, wearable or implantable electronic devices (e.g., pacemakers, neurostimulators, and defibrillators) also need high pulsed power supply, which is hard to deal with fuel cell alone. In addition, fuel cell needs a constant supply of reactants, including fuels and oxidants, to keep electricity production continuously, and if the reactants are scarce, the fuel cell will not work properly. These deficiencies can be remedied by the integration of supercapacitors with high power density. Thus, fuel cell with an internal energy store could represent a clear improvement.\(^{109}\)

To realize the all-in-one device, it is a common idea to modify the electrode with capacitive material. At the beginning, Deeke et al. modified the anode of the microbial fuel cell with activated carbon. The results show that device with capacitive bioanode has a significant improvement in performance and storage capacity compared with pure fuel cell.\(^{110}\) Later, in 2013, Pankratov et al. proposed the concept of self-charging electrochemical biocapacitor, which combines enzyme fuel cell and supercapacitor for the first time. As shown in Figure 7A, the PANI/carbon nanotube composites loaded on one side of the graphite foil and gold nanoparticles with two redox enzymes catalysts loaded on the other side. The device improves power output significantly by a factor of 170 in comparison to state-of-the-art enzyme fuel cells at that time.\(^{111}\) They then applied the same charge storage material into ascorbate-oxygen fuel cell and obtained similar results with energy density and power density of 0.5 Wh/kg and 23 W/kg, respectively.\(^{112}\) Around the same time, a similar concept of glucose/O\(_2\) fuel cell with dual-function electrodes is proposed. As shown in Figure 7B, the anode with carbon nanotubes wiring of glucose oxidase can not only oxidize glucose, but also store electrons; while the cathode with carbon nanotubes wiring of laccase can not only reduce oxygen, but also store holes.\(^{113}\) The device can achieve constantly 2 mW per discharge pulse for more than 40 000 pulses, which is sufficient to run a pacemaker. It is possible to power implantable electronics like actuator and defibrillator with further optimization. It is worth noting
that these devices still have defects in terms of long-term and operational stability. The material selection, the ratio of energy storage materials, and catalytic materials, as well as the electrode interface need to be further optimized. In spite of this, the value of these works is to put forward the hybrid electric power device for simultaneous generation and storage of electric energy, thus, providing reference for future research.\textsuperscript{114}

At present, the energy storage materials involved in all-in-one devices with combination of enzyme biofuel cell and supercapacitor mainly include carbon materials, metal nanoparticles, and conductive polymers.\textsuperscript{115–117} Moreover, one redox polymer of poly(vinyl imidazole-co-allylamine)[Os(bpy)\textsubscript{2}Cl] is introduced to fabricate the first “Nernstian biosupercapacitor.” The oxidation of glucose at the anode and the reduction of oxygen at the cathode will induce the redox reaction of Os\textsuperscript{3+}/Os\textsuperscript{2+}, making the Nernst potential of anode higher than that of cathode. Despite the use of one and the same redox mediator on both cathodic and anodic sides, a large power density up to eight times higher than that of the corresponding biofuel cells is still obtained for a short time.\textsuperscript{118} And MnO\textsubscript{2} can be directly used...
as cathode material and assembled with capacitive biological anode to form an oxygen-free and membrane-less all-in-one device due to its suitable discharge potential and inert to glucose. In addition, spontaneous potential recovery can be achieved through the redistribution of oxidation state when only a fraction is discharged in pulse mode. The device eliminates the oxygen shortage of conventional biofuel cells and is expected to be used as an intermittent power source for implanted medical devices. To better carry out implantable application research, it is necessary to consider the biocompatibility and biodegradability of materials. To solve this problem, biological protein molecules with their own redox capabilities are introduced into the system as charge storage materials. At the same time, the presence of the protein also promotes the charge transfer between the glucose dehydrogenase and electrode, significantly increasing the power density to 4.5 MW/cm² in pulse mode. To further demonstrate the practical application of the system, the first sweat-based wearable and scalable all-in-one device is prepared, which can capture and store energy from sweat lactate and deliver high power pulses. As shown in Figure 7C, the epidermal device combines carbon nanotube ink with electrodeposited polypyrrole on the anode and porous platinum on the cathode to enable bioenergy conversion and rapid self-charging from sweat produced by human activity. The device not only has high pulse output power and stable cycling performance in the case of sweating, but also can output high power pulse for a period of time after sweating stops due to its charge storage function. The device with high power of 1.7 mW/cm² in vitro and even 343 μW/cm² on the body during exercise exhibits great potential as a power source for the next generation of wearable electronics.

In addition, high-performance integrated hybrid energy devices based on proton exchange membrane fuel cell are rarely reported. Sun et al. achieved the organic unification of energy conversion and storage by redesigning traditional electrodes in Figure 7D. The pseudocapacitive material of PANI nanowire arrays is first constructed on a carbon cloth, and then the catalysts that can catalyze oxygen reduction reaction and methanol reduction reaction are sprayed onto the PANI electrodes, respectively. In the open circuit, the PANI in cathode is oxidized with the charge transferred to oxygen, while the PANI in anode is reduced with methanol reduction, thus, realizing the self-charging process. A good match between the pseudocapacitive potential of charge and discharge process and electrocatalytic reaction potential makes the device self-charging. In conclusion, the dual-function electrodes based on PANI organically combine the electrocatalytic reaction of proton exchange membrane fuel cell and the pseudocapacitance charge-discharge process of supercapacitor. Benefitting from this, the all-in-one device achieves remarkably specific energy density of 1550 Wh/kg and power density of 4080 W/kg. The superior performance will facilitate its future roles in electric vehicles, smart grid energy storage, and consumer electronics. The electrochemical interface between the capacitive material and the catalyst is optimized to improve the charge transfer, thus, further improving the performance. The strategy of electrochemical interfaces customization also provides other avenues for performance improvement.

In other words, the all-in-one power device integrating the energy harvesting function of fuel cell with high energy density and the energy storage function of supercapacitor with high power operation has seen rapid development in recent decades. As shown in Figure 7E, the devices employing dual-function electrodes for charge storing and catalysis in place of traditional single-function anode and cathode. The materials used for catalysis include noble metals, nonmetallic catalysts, and biological-related materials (e.g., enzymes, redox proteins, membrane-resident redox entities, organelles, as well as whole living cells), while the materials for charge storage cover carbon nanomaterials, metal oxide, conducting polymer, and biological materials. In this system, high power output is expected and achieved by self-charging process. The self-charging operation mechanism is the fuel oxidation reaction and oxidant reduction reaction that occur at the anode and cathode, respectively. The electrons or holes generated in the reaction process will polarize the electrode, making it positively or negatively charged. The charged electrodes further attract the counter ions in the electrolyte to form double electric layer or induce the corresponding redox reaction within electrodes. The integrated system has the advantages of increased power density and operational flexibility compared to pure fuel cell and supercapacitor.

### 3.5 All-in-one power system based on other energy

In addition to the above-mentioned all-in-one power system, the integrated energy system utilizing streaming potential via fluid flow in the charged narrow nanochannel has been reported. A typical example is the electokinetic self-charging supercapacitor first designed in Figure 8A. The anodic aluminum oxide nanochannels membrane as separator and carbon nanotube as electrode are configured together. As shown in Figure 8B, when the electrolyte of NaCl solution flows through the narrow positively charged nanochannels under the action of external pressure, the positive and negative ions in the electrolyte will form asymmetrical distribution at both ends of the nanochannels. There will be more Na⁺ ions on the top layer and more Cl⁻ ions on the bottom layer, resulting in
a potential difference between the two electrodes. When the two electrodes are connected through external circuit, electrons are transferred to balance the potential difference created by the streaming potential. The transferred charges are stored in the carbon nanotube electrode by forming double layer, which can eventually be released when needed. The device is able to store a charge density of 0.4 mC/cm² under 2.5 bar. Although the performance of the device is mediocre, the subsequent optimization of electrode materials and separator is expected to further improve the performance. The device shows potential applications in microelectronics and micro-/nanofluids through the utilization of a wide range of mechanical vibrations and forces. Electrolyte flow can be driven by not only external pressure, but also by gravity everywhere on Earth.¹²⁶ In conclusion, it is an innovative attempt to realize the integrated construction of energy devices depending on streaming potential, which provides a reference for more research.

### 3.6 Perspectives

To sum up, this section summarizes the all-in-one power system based on different energy sources such as mechanical, thermal, solar, and chemical energy, and so on. The different power systems have their own unique characteristics and face specific challenges. Considering the huge differences in structure design and working environment of each unit, the design of all-in-one power system is much more difficult than the integration mode of external circuits connecting independent units. In essence, the all-in-one power system is an energy conversion device with energy storage function or energy storage device with energy conversion function. Through the introduction and analysis of the all-in-one power system, we summarize two kinds of self-charging strategies. The first one is shown in Figure 9A, cations and anions in the electrolyte migrate toward two electrodes, respectively, under the direct external stimulation or the external stimulation induced internal electric field to form a potential difference, which will further induce the electrode to produce double electric layers or redox reactions. When the external stimulus is removed, the ion potential difference disappears and the energy stored in the electrode is revealed, thus, device is charged (Figure 9B). This mode is usually used to charge the device under the stimulation of mechanical external force and temperature difference. The second one in Figure 9C which shows that the two dual-function electrodes undergo different redox reactions, respectively, under external stimulation. The generated electrons or holes will induce the electrode to undergo energy storage reaction, and eventually part of the electric energy will be stored in the electrodes. The reactions on the electrodes disappear after the external stimulus is absent, and the electrical energy stored in the electrode comes into play (Figure 9D). The second strategy is often used to collect and store solar or chemical energy simultaneously.
4 | CONCLUSIONS AND PERSPECTIVES

The integrated system of energy conversion and storage devices is of great significance to the development of next-generation power systems since the integrated system can solve some defects of the individual energy conversion or storage device unit. The integrated system has developed from two independent devices connected by external circuits or sharing a single electrode or the planar integration system into all-in-one system through the efforts of recent decades. Compared with the previous three integration modes, all-in-one power system with higher degree of integration possesses its own unique advantages. Generally speaking, the all-in-one power system is more difficult to design and less difficult to manufacture.

Despite aforementioned impressive conceptual breakthroughs, the integrated power systems with unsatisfactory performance still have a long way to go before they can significantly reduce costs and improve efficiency and stability. Thus, innovative approaches are still needed to develop highly integrated power systems with better performances and wider application prospects. We summarize some perspectives and opportunities for future research directions here.

First, the self-charging and self-storage mechanisms need further understanding. The research on the mechanism of independent energy conversion and storage devices will directly promote studies on integrated systems. The performance matching between two units will have a great impact on charging process, so attention should be paid to the matching problem to prevent inefficiency caused by unequal performance. Current understanding regarding the self-charging process is that there are still many problems and controversies in all-in-one power systems exist. Most studies prove the synchronous conversion and storage of energy through the final experimental results, but there is a lack of effective data to support the explanation of ion and electron migration and storage involved in the process. In the future, more in-situ tests should be carried out to assist in explaining the correctness of the mechanism. In addition, a simple, typical, ideal model can be attempted to establish mechanism exploration alone. Molecular dynamics simulation, numerical simulation, and other methods should be fully used to simulate and calculate the ion migration and electron storage behaviors in these experimental processes, so as to deepen the research on the mechanism. Second, performance improvements are the driving forces of expanding application scenarios and advancing the field. At present, the performance of the highly integrated power system is not satisfactory and generally lower than that of the corresponding device system connected by external circuits. The all-in-one power devices are generally in the proof-of-concept and are moving toward the performance optimization stage through material optimization and structural innovation, so as to break through more application barriers by greatly improving performance. Third, high integration remains one of the key challenges for future device development. The integration forms involved in this review are relatively simple with just the combination of one type of energy conversion and storage devices. More complex integration systems should be explored, such as the combination of different types of power generator and storage devices. For example, the combination system of triboelectric nanogenerator and the solar cell can collect the energy by absorbing sunlight on sunny days and vibration of raindrops on rainy days. This multienergy conversion method is an effective supplement to the existing single energy conversion system, and greatly improves the utilization rate of energy in the environment by collecting and converting more energy. Moreover, it is necessary to integrate energy devices with electric equipment and devices. For example, the advent of the Internet of Things will require energy systems to power widely distributed sensing systems, thus, the integration of power system and sensing system is of great value in the field of self-powered system. Highly integrated systems, including energy conversion, storage, and functional devices, also directly expand their application scenarios. The comprehensive system has great application potential not only in wearable electronic devices, but also in remote monitoring, personalized health monitoring, and industrial and agricultural intelligent management. Fourth, the reported all-in-one systems are generally presented as macroscopic 2D and 3D devices, which limit the compatibility with portable and wearable electronics. The miniaturization and planarization of all-in-one systems will significantly improve this problem and provide more opportunities for wider application. Fifth, special functions, such as antideformation, self-healing, and self-cleaning, should also be added to the energy system to adapt to more extreme scenarios. In summary, with increasing interest in this field, materials scientists, chemists, and electronics specialists will work together to develop a highly integrated power system with excellent performance. And the all-in-one power system, as an interdisciplinary research field, will make rapid and huge progress with the help of the improvement and development of fields including electronic integration, energy conversion, and energy storage.

ACKNOWLEDGMENTS

This work was financially supported by the NSFC (22075019, 22035005) and the National Key R&D Program of China (2017YFB1104300).
CONFLICT OF INTEREST

The authors declare no conflict of interest.

ORCID

Changxiang Shao https://orcid.org/0000-0001-8476-8516
Yang Zhao https://orcid.org/0000-0002-8187-9963
Liangti Qu https://orcid.org/0000-0002-7320-2071

REFERENCES

1. Chu S, Majumdar A. Opportunities and challenges for a sustainable energy future. Nature. 2012;488:294-303.
2. Dresselhaus MS, Thomas IL. Alternative energy technologies. Nature. 2001;414(6861):332-337.
3. Lund H. Renewable energy strategies for sustainable development. Energy. 2007;32(6):912-919.
4. Levenda AM, Behrsin I, Disano F. Renewable energy for whom? A global systematic review of the environmental justice implications of renewable energy technologies. Energy Research & Social Science. 2021;71:101837.
5. Leijtens T, Bush KA, Prasanna R, et al. Opportunities and challenges for tandem solar cells using metal halide perovskite semiconductors. Nat Energy. 2018;3:828-838.
6. Tai Q, Yan F. Emerging semitransparent solar cells: materials and device design. Adv Mater. 2017;29(34):1700192.
7. O’Regan B, Grätzel M. A low-cost, high-efficiency solar cell based on dye-sensitized colloidal TiO2 films. Nature. 1991;353(6346):737.
8. Wang Z, Song J. Piezoelectric nanogenerators based on zinc oxide nanowire arrays. Science. 2006;312(5771):242-246.
9. Liu Z, Nie J, Miao B, et al. Self-powered intracellular drug delivery by a biomechanical energy-driven triboelectric nanogenerator. Adv Mater. 2019;31(12):1807795.
10. Zhao X, Kuang S, Wang Z, et al. Highly adaptive solid-liquid interfacing triboelectric nanogenerator for harvesting diverse water wave energy. ACS Nano. 2018;12(5):4280-4285.
11. Xu M, Wang P, Wang Y, et al. A soft and robust spring based triboelectric nanogenerator for harvesting arbitrary directional vibration energy and self-powered vibration sensing. Adv Energy Mater. 2018;8(9):1702432.
12. Siddiqui S, Lee HB, Kim DI, et al. An omnidirectionally stretchable piezoelectric nanogenerator based on hybrid nanofibers and carbon electrodes for multimodal straining and human kinematics energy harvesting. Adv Energy Mater. 2018;8(2):1701520.
13. Choi J, Jung Y, Yang SJ, et al. Flexible and robust thermoelectric generators based on all-carbon nanotube yarn without metal electrodes. ACS Nano. 2017;11(8):7608-7614.
14. Wang X, Tan C, Chan K, et al. In-built thermo-mechanical cooperative feedback mechanism for self-propelled multimodal locomotion and electricity generation. Nat Commun. 2018;9:3438.
15. Im H, Kim T, Song H, et al. High-efficiency electrochemical thermal energy harvester using carbon nanotube aerogel sheet electrodes. Nat Commun. 2016;7:10600.
16. Jung YS, Jeong DH, Kang SB, et al. Wearable solar thermoelectric generator driven by unprecedentedly high temperature difference. Nano Energy. 2017;40:663-672.
17. Lee S, Kim K, Kang D-H, et al. Vertical silicon nanowire thermoelectric modules with enhanced thermoelectric properties. Nano Lett. 2019;19(2):747-755.
18. Chen X, Goodnight D, Gao Z, et al. Scaling up nanoscale water-driven energy conversion into evaporation-driven engines and generators. Nat Commun. 2015;6:7346.
19. Huang Y, Cheng H, Yang C, et al. Interface-mediated hygro-electric generator with an output voltage approaching 1.5 volts. Nat Commun. 2018;9(1):4166.
20. Xue G, Xu Y, Ding T, et al. Water-evaporation-induced electricity with nanostructured carbon materials. Nat Nanotechnol. 2017;12(4):317-321.
21. Wang H, Sun Y, He T, et al. Bilayer of polyelectrolyte films for spontaneous power generation in air up to an integrated 1000 V output. Nat Nanotechnol. 2021;16:811-819.
22. Xu Y, Chen P, Zhang J, et al. A one-dimensional fluidic nanogenerator with a high power conversion efficiency. Angew Chem Int Ed. 2017;56(42):12940-12945.
23. Kim SH, Haines CS, Li N, et al. Harvesting electrical energy from carbon nanotube yarn twist. Science. 2017;357(6353):773-778.
24. Xu T, Ding X, Huang Y, et al. An efficient polymer moist-electric generator. Energy Environ Sci. 2019;12(3):972-978.
25. Zhang Z, Li X, Yin J, et al. Emerging hydrovoltaic technology. Nat Nanotechnol. 2018;13(12):1109.
26. Liu X, Gao H, Ward JE. Power generation from ambient humidity using protein nanowires. Nature. 2020;578:550-554.
27. Ryu H, Yoon HJ, Kim W. Hybrid energy harvesters: toward sustainable energy harvesting. Adv Mater. 2019;31(34):1802898.
28. Zhu Q, Zhao D, Chen M, et al. A new view of supercapacitors: integrated supercapacitors. Adv Energy Mater. 2019;9(36):1901081.
29. Fleischmann S, Mitchell JB, Wang R, et al. Pseudocapacitance: from fundamental understanding to high power energy storage materials. Chem Rev. 2020;120(14):6738-6782.
30. Whittingham MS. Introduction: batteries. Chem Rev. 2014;114(23):11413.
31. Huang Y, Zeng Y, Yu M, et al. Recent smart methods for achieving high-energy asymmetric supercapacitors. Small Methods. 2018;2(2):1700230.
32. Dai C, Sun G, Hu L, et al. Recent progress in graphene-based electrodes for flexible batteries. InfoMat. 2020;2:509-526.
33. Ye M, Zhang Z, Zhao Y, et al. Graphene platforms for smart energy generation and storage. Joule. 2018;2(2):245-268.
34. Ji B, Chen N, Shao C, et al. Intelligent multiple-liquid evaporation power generation platform using distinctive Jaboticablike carbon nanosphere@TiO2 nanowires. J Mater Chem A. 2019;7(12):6766-6772.
35. Yoon SG, Jin H, Lee WH, et al. Evaporative electrical energy generation via diffusion-driven ion-electron-coupled transport in semiconducting nanoporous channel. Nano Energy. 2021;80:105522.
36. Gao J, Shao C, Shao S, et al. Laser-assisted large-scale fabrication of all-solid-state asymmetrical micro-supercapacitor array. Small. 2018;14(37):1801809.
37. Gao C, Gao J, Shao C, et al. Versatile origami micro-supercapacitors array as a wind energy harvester. J Mater Chem A. 2018;6(40):19750-19756.
38. Qin S, Qian Z, Yang X, et al. Hybrid piezo/triboelectric-driven self-charging electrochromic supercapacitor power package. Adv Energy Mater. 2018;8(23):1800069.

39. Chai Z, Zhang N, Sun P, et al. Tailorable and wearable textile devices for solar energy harvesting and simultaneous storage. ACS Nano. 2016;10(10):9201-9207.

40. Zhang H, Cao Y, Chee MOL, et al. Recent advances in micro-supercapacitors. Nanoscale. 2019;11(13):5807-5821.

41. Lu B, Jin X, Han Q, et al. Planar graphene-based microsupercapacitors. Small. 2021;17(48):2006827.

42. Wang Y, Zhao Y, Qu L. Laser fabrication of functional micro-supercapacitors. J Energy Chem. 2021;59:642-665.

43. Jing X, Song L, Dai C, et al. An aqueous anti-freezing and heat-tolerant symmetric microsupercapacitor with 2.3 V output voltage. Adv Energy Mater. 2021;11(33):2101523.

44. Jing X, Zhang G, Sun G, et al. Flexible and high-performance microsupercapacitors with wide temperature tolerance. Nano Energy. 2019;64:103938.

45. Sun G, Yang H, Gu Zhang, et al. A capacity recoverable zinc-ion micro-supercapacitor. Energy Environ Sci. 2018;11:3367-3374.

46. Gao C, Bai C, Gao J, et al. A directly swallowable and ingestible micro-supercapacitor. J Mater Chem A. 2020;8(7):4055-4061.

47. Liu Q, Zhang G, Chen N, et al. The first flexible dual-ion microbattery demonstrates superior capacity and ultra-high energy density: small and powerful. Adv Funct Mater. 2020;30(38):2002086.

48. Sun G, Jin X, Yang H, et al. Aqueous Zn-MnO2 rechargeable microbattery. J Mater Chem A. 2018;6(23):10926-10931.

49. Gao C, Huang J, Xiao Y, et al. A seamlessly integrated device of micro-supercapacitor and wireless charging with ultra-high energy density and capacitance. Nat Commun. 2021;12:2647.

50. Zhao F, Cheng H, Zhang Z, et al. Direct power generation from a graphene oxide film under moisture. Adv Mater. 2015;27:4351-4357.

51. Cheng H, Huang Y, Qu L, et al. Flexible in-plane graphene oxide moisture-electric converter for touchless interactive panel. Nano Energy. 2018;45:37-43.

52. Huang Y, Cheng H, Shi G, et al. Highly efficient moisture-triggered nanogenerator based on graphene quantum dots. ACS Appl Mater Interfaces. 2017;9(44):38170-38175.

53. Yang C, Huang Y, Cheng H, et al. Rollable, stretchable, and reconfigurable graphene hydroelectric generators. Adv Mater. 2019;31(2):1805705.

54. Yang K, Cho K, Yang S, et al. A laterally designed all-in-one energy device using a thermoelectric generator-coupled micro supercapacitor. Nano Energy. 2019;60:667-672.

55. Qiu M, Sun P, Cui G, et al. A flexible, micro-supercapacitor with integral photocatalytic fuel cell for self-charging. ACS Nano. 2019;13(7):8246-8255.

56. Xue J, Wu H, Hu L, et al. Integrated photo-supercapacitor based on bi-polar TiO2 nanotube arrays with selective one-side plasma-assisted hydrogenation. Adv Funct Mater. 2014;24(13):1840-1846.

57. Zhou Y, Zhang S, Ding Y, et al. Efficient solar energy harvesting and storage through a robust photocatalyst driving reversible redox reactions. Adv Mater. 2018;30(31):1802294.

58. Guo W, Xue X, Wang S, et al. An integrated power pack of dye-sensitized solar cell and Li battery based on double-sided TiO2 nanotube arrays. Nano Lett. 2012;12(5):2520-2523.

59. Liu R, Takakuwa M, Li A, et al. An efficient ultra-flexible photo-charging system integrating organic photovoltaics and supercapacitors. Adv Energy Mater. 2020;10(20):2000523.

60. Han Y, Lu B, Shao C, et al. A hygroelectric power generator with energy self-storage. Chem Eng J. 2019;384:123366.

61. Zhang Z, Chen X, Chen P, et al. Integrated polymer solar cell and electrochemical supercapacitor in a flexible and stable fiber format. Adv Mater. 2014;26(3):466-470.

62. Xue X, Wang S, Guo W, et al. Hybridizing energy conversion and storage in a mechanical-to-electrochemical process for self-charging power cell. Nano Lett. 2012;12(9):5048-5054.

63. Xue X, Ping D, Shuang Y, et al. CuO/PVDF nanocomposite anode for a piezo-driven self-charging lithium battery. Energy Environ Sci. 2013;6(9):2615-2620.

64. Xue X, Ping D, He B, et al. Flexible self-charging power cell for one-step energy conversion and storage. Adv Energy Mater. 2014;4(5):1301329.

65. Kim Y, Xie Y, Wen X, et al. Highly porous piezoelectric PVDF membrane as effective lithium ion transfer channels for enhanced self-charging power cell. Nano Energy. 2015;14:77-86.

66. He H, Fu Y, Zhao T, et al. All-solid-state flexible self-charging power cell basing on piezo-electrolyte for harvesting/storing body-motion energy and powering wearable electronics. Nano Energy. 2017;39:590-600.

67. Zhou D, Xue L, Wang L, et al. Self-chargeable sodium-ion battery for soft electronics. Nano Energy. 2019;61:435-441.

68. Ramadoss A, Saravanakumar B, Lee SW, et al. Piezoelectric-driven self-charging supercapacitor power cell. ACS Nano. 2015;9(4):4337-4345.

69. Song R, Jin H, Li X, et al. A rectification-free piezo-supercapacitor with a polyvinylidene fluoride separator and functionalized carbon cloth electrodes. J Mater Chem A. 2015;3(29):14963-14970.

70. Parida K, Bhavanasi V, Kumar V, et al. Fast charging self-powered electric double layer capacitor. J Power Sources. 2017;342(28):70-78.

71. Maitra A, Karan SK, Paria S, et al. Fast charging self-powered wearable and flexible asymmetric supercapacitor power cell with fish swim bladder as an efficient natural bio-piezoelectric separator. Nano Energy. 2017;40:633-645.

72. Pazhamalai P, Krishnamoorthy K, Mariappan VK, et al. A high efficacy self-charging MoS2 solid-state supercapacitor using electrospun nanofibrous piezoelectric separator with ionogel electrolyte. Adv Mater Interfaces. 2018;5(12):1800055.

73. Sahoo S, Krishnamoorthy K, Pazhamalai P, et al. High performance self-charging supercapacitors using a porous PVDF-ionic liquid electrolyte sandwiched between two-dimensional graphene electrodes. J Mater Chem A. 2019;7(38):21693-21703.

74. Zhou D, Wang N, Yang T, et al. A piezoelectric nanogenerator promotes highly stretchable and self-chargable supercapacitors. Mater Horiz. 2020;7(8):2158-2167.

75. Zhou D, Wang F, Zhao X, et al. Self-chargable flexible solid-state supercapacitors for wearable electronics. ACS Appl Mater Interfaces. 2020;12(40):44883-44891.

76. Rasheed A, He W, Qian Y. A flexible supercapacitor type rectifier-free self-charging power unit based on a multifunctional PVDF-ZnO-RGO piezoelectric matrix. ACS Appl Mater Interfaces. 2020;12(18):20891-20900.

77. Manoharan S, Pazhamalai P, Mariappan VK, et al. Proton conducting solid electrolyte-piezoelectric PVDF hybrids: novel
bifunctional separator for self-charging supercapacitor power cell. *Nano Energy*. 2021;83:105753.

78. Krishnamoorthy K, Pazhamalai P, Mariappan VK, et al. Probing the energy conversion process in piezoelectric-driven electrochemical self-charging supercapacitor power cell using piezoelectrochemical spectroscopy. *Nat Commun*. 2020;11(1):2351.

79. Mentor JJ, Torres R, Hallinan DT. The Soret effect in dry polymer electrolyte. *Mol Syst Des Eng*. 2020;5(4):856-863.

80. Dan Z, Fabiano S, Berggren M, et al. Ionic thermoelectric gating organic transistors. *Nat Commun*. 2017;8:14214.

81. Al-zubaidi A, Ji X, Yu J. Thermal charging of supercapacitors: a perspective. *Sustain Energy Fuels*. 2017;1(7):1457-1474.

82. Wang H, Zhao D, Khan ZU, et al. Ionic thermoelectric figure of merit for charging of supercapacitors. *Adv Electron Mater*. 2017;3(4):1700013.

83. Zhao D, Wang H, Khan ZU, et al. Ionic thermoelectric supercapacitors. *Energy Environ Sci*. 2016;9(4):1450-1457.

84. Arpan K, Fisher TS. Harnessing the thermogalvanic effect of the ferro/ferricyanide redox couple in a thermally chargeable supercapacitor. *Electrochem Acta*. 2018;281:357-369.

85. Kim SL, Lin H, Yu C. Thermally chargeable solid-state supercapacitor. *Adv Energy Mater*. 2016;6(18):1600546.

86. Kim SL, Hsu JH, Yu C. Intercalated graphene oxide for flexible and practically large thermoelectric voltage generation and simultaneous energy storage. *Nano Energy*. 2018;48:582-589.

87. Wu X, Huang B, Wang Q, et al. Thermally chargeable supercapacitor using a conjugated conducting polymer: insight into the mechanism of charge-discharge cycle. *Chem Eng J*. 2019;373:493-500.

88. Ding Y, Guo X, Katrina R, et al. Simultaneous energy harvesting and storage via solar-driven regenerative electrochemical cycles. *Energy Environ Sci*. 2019;12(11):3370-3379.

89. Luo B, Ye D, Wang L. Recent progress on integrated energy conversion and storage systems. *Adv Sci*. 2017;4(9):1700104.

90. Zeng Q, Lai Y, Jiang L, et al. Integrated photorechargeable energy storage system: next-generation power source driving the future. *Adv Energy Mater*. 2020;10(14):1903930.

91. Miyasaka T, Murakami TN. The photocapacitor: an efficient self-charging capacitor for direct storage of solar energy. *Appl Phys Lett*. 2004;85(17):3932.

92. Takshi A, Yaghoubi H, Tevi T, et al. Photoactive supercapacitors for solar energy harvesting and storage. *J Power Sources*. 2015;275:621-626.

93. Safshekan S, Herraiz-Cardona I, Cardenas-Morcoso D, et al. Solar energy storage by a heterostructured BiVO₄-PbO₂ photocapacitive device. *ACS Energy Lett*. 2017;2(2):469-475.

94. Liu D, Zl W, Sajjad SD, et al. Reversible electron storage in an all-vanadium photoelectrochemical storage cell: synergy between vanadium redox and hybrid photocatalyst. *ACS Catal*. 2015;5(4):2632-2639.

95. Wang P, Chen X, Sun G, et al. A capacitor-type faradaic junction for direct solar energy conversion and storage. *Angew Chem Int Ed*. 2021;60(3):1390-1395.

96. Wang Y, Tang J, Peng Z, et al. Fully solar-powered photoelectrochemical conversion for simultaneous energy storage and chemical sensing. *Nano Lett*. 2014;14(6):3668-3673.

97. Podjaski F, Kröger J, Lotsch BV. Toward an aqueousolar battery: direct electrochemical storage of solar energy in carbon nitrides. *Adv Mater*. 2018;30(9):1705477.

98. Ren Y, Zhu T, Liu Y, et al. Direct utilization of photoinduced charge carriers to promote electrochemical energy storage. *Small*. 2021;17(21):2008047.

99. Zhu M, Huang Y, Huang Y, et al. Capacitance enhancement in a semiconductor nanostructure-based supercapacitor by solar light and a self-powered supercapacitor-photodetector system. *Adv Funct Mater*. 2016;26(25):4481-4490.

100. An C, Wang Z, Xi W, et al. Nanoporous Cu@Cu₂O hybrid arrays enable photoassisted supercapacitor with enhanced capacities. *J Mater Chem A*. 2019;7(26):15691-15697.

101. Wang Q, Chen H, McFarland E, et al. Solar rechargeable batteries based on lead-organohalide electrolyte. *Adv Energy Mater*. 2015;5(24):1501418.

102. Hauch A, Georg A, Kras’ovec UO, et al. Photovoltaically self-charging battery. *J Electrochem Soc*. 2020;149(9):A1208-A1211.

103. Xu C, Zhang X, Duan L, et al. A photo-assisted rechargeable battery: synergy, compatibility, and stability of a TiO₂/dye/Cu₂S bifunctional composite electrode. *Nanoscale*. 2020;12(2):530-537.

104. Lv J, Tan Y, Xie J, et al. Direct solar-to-electrochemical energy storage in a functionalized covalent organic framework. *Angew Chem Int Ed*. 2018;57(39):12716-12720.

105. Li J, Ren C, Zhang L, et al. Hybridized S cathode with N719 dye for a photo-assisted charging Li-S battery. *J Energy Chem*. 2022;65:205-209.

106. Nikiforidis G, Tajima K, Byon HR. High energy efficiency and stability for photoassisted aqueous lithium-iodine redox batteries. *ACS Energy Lett*. 2016;1(4):806-813.

107. Yu M, McCulloch WD, Beauchamp DR, et al. Aqueous lithium-iodine solar flow battery for the simultaneous conversion and storage of solar energy. *J Am Chem Soc*. 2015;137(26):8332-8335.

108. Liu X, Yuan Y, Liu J, et al. Utilizing solar energy to improve the oxygen evolution reaction kinetics in zinc-air battery. *Nat Commun*. 2019;10:4767.

109. Pankratov V, Bollenga P, Pankratov D, et al. Supercapacitive biofuel cells. *Curr Opin Biotech*. 2022;73:1-9.

110. Deeke A, Sleutels THJA, Hamelers HVM, et al. Capacitive bioanodes enable renewable energy storage in microbial fuel cells. *Environ Sci Technol*. 2012;46:3554-3560.

111. Pankratov D, Blum Z, Suyatin DB, et al. Self-charging electrochemical biocapacitor. *ChemElectroChem*. 2014;1(2):343-346.

112. Pankratov D, Falkman P, Blum Z, et al. A hybrid electric power device for simultaneous generation and storage of electric energy. *Energy Environ Sci*. 2014;7(3):989-993.

113. Agn’e C, Holzinger M, Goff AL, et al. Supercapacitor/biofuel cell hybrids based on wired enzymes on carbon nanotube matrices: autonomous reloading after high power pulses in neutral buffered glucose solutions. *Energy Environ Sci*. 2014;7(6):1884-1888.

114. Pankratov D, Blum Z, Shleev S. Hybrid electric power bio-devices. *ChemElectroChem*. 2014;1(11):1798-1807.

115. Knoche KL, Hickey DP, Milton RD, et al. Hybrid glucose/O₂ biobattery and supercapacitor utilizing a pseudocapacitive dimethylferrocene redox polymer at the bioanode. *ACS Energy Lett*. 2016;1(2):380-385.

116. Xiao X, Conghaile PÓ, Leech D, et al. A symmetric supercapacitor/biofuel cell hybrid device based on enzyme-modified nanoporous gold: an autonomous pulse generator. *Biosens Bioelectron*. 2017;90:96-102.
AUTHOR BIOGRAPHIES

Changxiang Shao received her PhD degree from the Beijing Institute of Technology in 2021. She is now an Associate Professor at the School of Chemistry and Pharmaceutical Engineering in Shandong First Medical University & Shandong Academy of Medical Sciences. Her research interests lie in the development of new energy conversion and storage devices based on nanomaterials.

Yang Zhao received her PhD degree in physical chemistry from the Beijing Institute of Technology (Beijing, China). She is now an Assistant Professor of Chemistry at the Beijing Institute of Technology. Her research interests lie in the development of new methods and strategies for the fabrication of carbon-based functional materials for various applications.

Liangti Qu received a PhD in Chemistry from Tsinghua University (Beijing, China) in 2004. He is now a professor in the Department of Chemistry at Tsinghua University and leads the nanocarbon research group. His research interests in materials chemistry mainly focus on the synthesis, functionalization, and application of nanomaterials with carbon–carbon conjugated structures including carbon nanotubes, graphene, and conducting polymers.

How to cite this article: Shao C, Zhao Y, Qu L. Recent advances in highly integrated energy conversion and storage system. SusMat. 2022;2:142–160. https://doi.org/10.1002/sus2.48