



## Review

## Advances in integrated self-charging capacitors and batteries

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## ABSTRACT

Energy storage technologies have enabled a wide range of applications, from portable power supplies to tiny sensors. However, the advancement of certain applications is hindered by environmental factors and reliance on external power sources. Developing integrated self-charging energy storage systems is therefore of paramount importance. This refined review summarizes recent advancements in integrated self-charging capacitors and batteries, focusing on three key mechanisms: physical piezoelectricity, chemical oxidation, and biological enzyme catalysis. We analyze the material systems selection and design exhibiting self-charging behavior, elucidate the underlying reaction mechanisms, and highlight the challenges and potential solutions in existing designs. Furthermore, we envision future applications of self-charging energy storage devices, which are expected to attract significant attention in physics, chemistry, and biological energy storage.

## 1. Introduction

High-power-density capacitors and high-energy-density lithium-ion batteries (LIBs) have become indispensable secondary power sources in various fields [1–3]. Nevertheless, environmental factors and the need for external charging can disrupt their operation, particularly in applications such as detection equipment, flexible electronics, and in vivo sensors, where uninterrupted functionality is critical. Recent advancements in materials science have led to the development of integrated self-charging systems that combine energy harvesting and storage within a single device, offering a promising solution to these persistent challenges [4–10].

The integrated self-charging batteries and capacitors convert mechanical, chemical, or biological energy into electrical energy, which is then stored as chemical energy [11–16]. Physical processes such as piezoelectricity, triboelectricity, and pyroelectricity are commonly employed for energy generation. Among these, piezoelectric-based integrated self-charging capacitors and LIBs are particularly promising for smart wearable electronics and sensors [17–34]. Chemical oxidation-based systems, utilizing air-oxidizable materials, are also gaining

traction for outdoor detection equipment [35–44]. In addition, biological enzyme-catalysis offers potential for in vivo health monitoring sensors [45–50]. Despite the progress, systematic reviews on self-charging energy storage are still lacking, necessitating a comprehensive analysis of material selection, reaction mechanisms, and applications.

This review focuses on recent advances in integrated self-charging capacitors and batteries, covering physical piezoelectricity, chemical oxidation, and biological enzyme catalysis (Fig. 1). We first summarize the material systems and working mechanisms underlying these self-charging technologies. Specifically, 1) Physical piezoelectric self-charging relies on piezoelectric materials, such as polyvinylidene fluoride (PVDF) based polymers, piezoelectric ceramics, and their composites. The piezoelectric effect, induced by mechanical stress on the piezoelectric film, drives the migration of electrolyte ions toward the positive and negative electrodes, where redox reactions occur. This process facilitates the conversion of mechanical energy into electrical energy, enabling self-charging. 2) Chemical oxidative self-charging utilizes spontaneously oxidizable materials, including vanadium-based, Prussian blue, polymers, and functionalized carbon materials as

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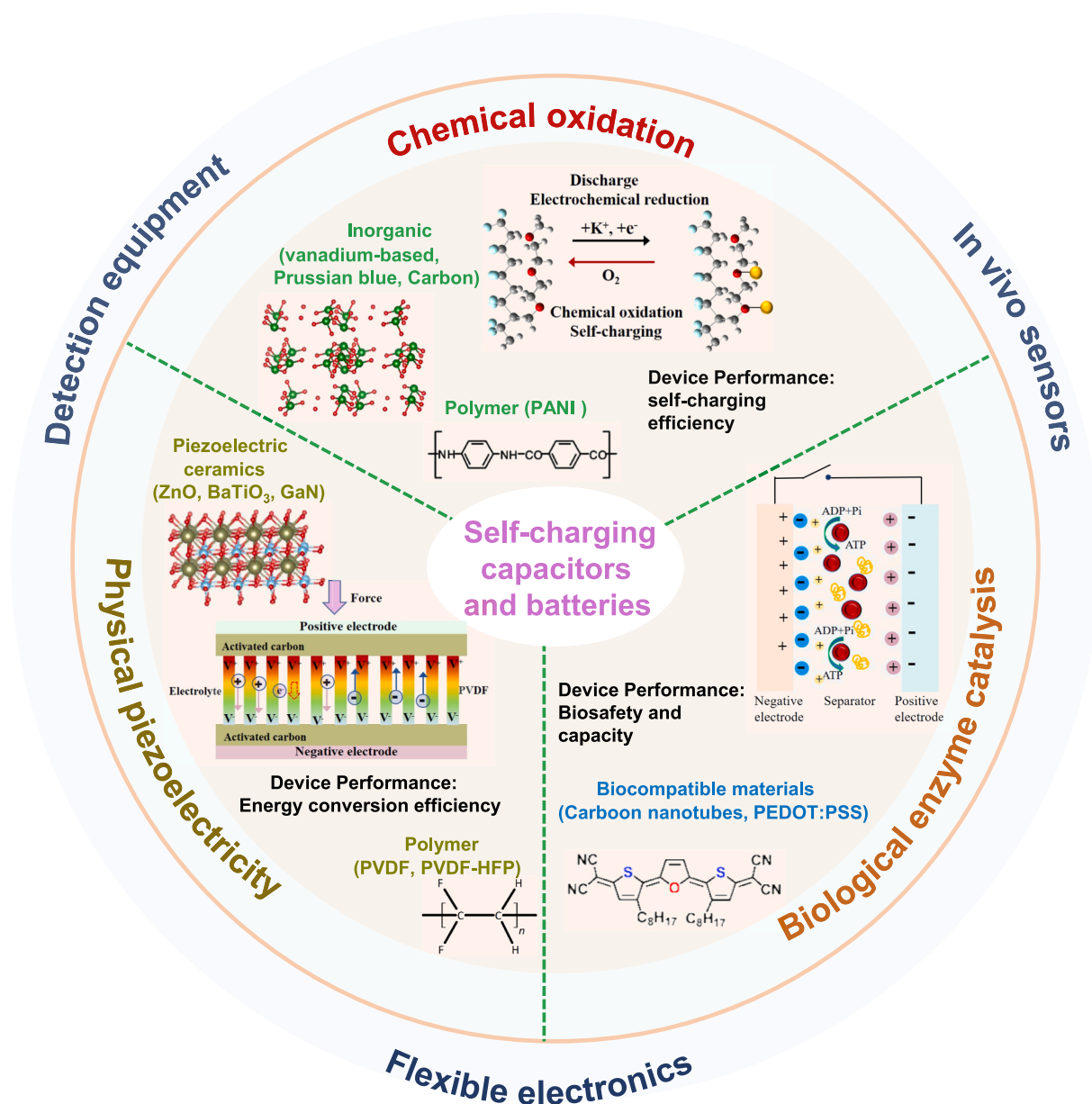


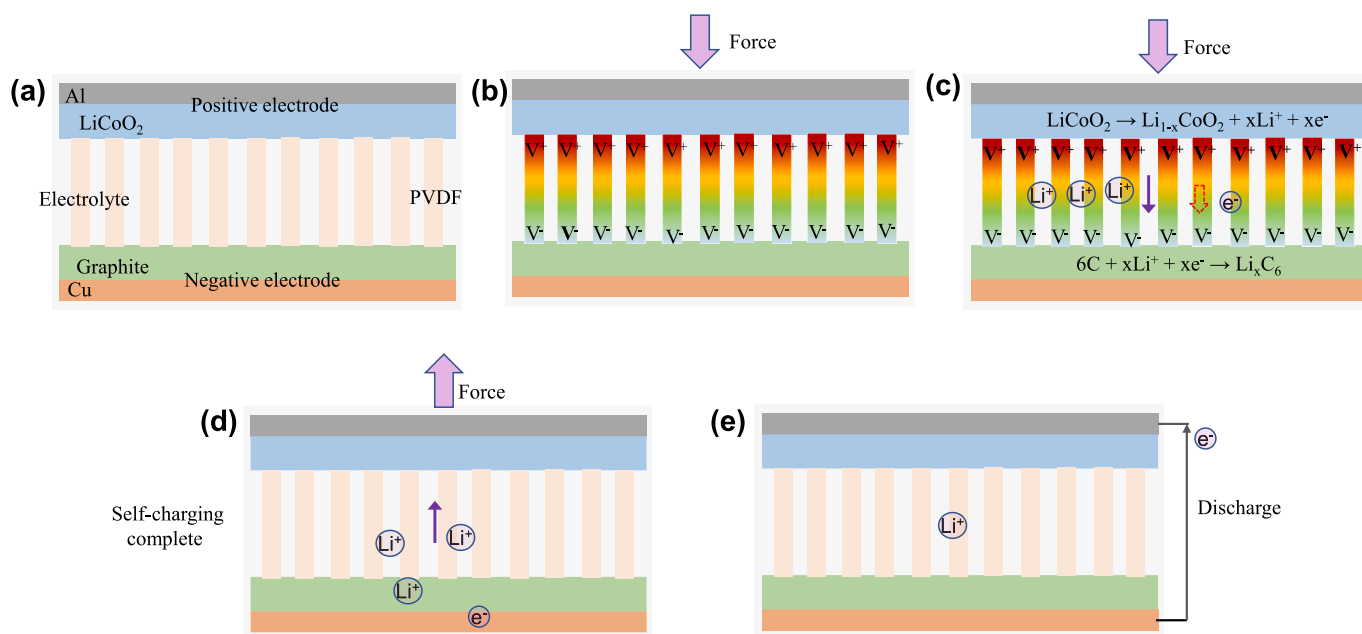
Fig. 1. Material, mechanism, and application of integrated self-charging capacitors and batteries.

electrode materials. During discharge, low-valence products are formed, which can be spontaneously oxidized to high-valence states by ambient oxygen, completing the self-charging cycle. 3) Biocompatible materials are employed in biological systems as components of bio-nanocapacitors. The self-charging process is driven by redox-active components within living organisms, such as metal centers in proteins, redox cofactors in enzymes, or cellular metabolism. Following this, we outline prospective research directions, emphasizing the most promising applications of self-charging nano-micro devices, particularly those related to the human body. These devices utilize bodily fluids (e.g., blood, lymph, and saliva) as the medium for energy harvesting and storage. Notable examples include nanoscale supercapacitors designed to power pH detectors in blood, cancer cell detectors in lymph, and devices that promote bone regeneration in the jaw by leveraging the piezoelectric effect in saliva. Throughout the review, we critically address the challenges and limitations of current self-charging designs,

offering insights into potential solutions. This review aims to the rapid and healthy development of self-charging energy storage.

## 2. Physical piezoelectric self-charging energy storage

Various approaches for converting mechanical energy into electrical energy include piezoelectricity, triboelectricity, and pyroelectricity. Among these, piezoelectric-based capacitors and batteries have emerged as the most feasible integrated self-charging devices [9]. The piezoelectric effect, first discovered by the Curie brothers in 1880, enables the conversion of mechanical energy into electrical energy, where the mechanical stress may be vibration, body movement, ultrasound, etc [51]. Building on this principle, Wang's group proposed piezoelectric nanogenerators (PENGs) by scanning ZnO nanowires with an atomic force microscope tip in 2006 [52]. The strain field-induced internal deformation of the ZnO nanowires resulted in charge separation and the



**Fig. 2.** The working mechanism of PENG-based self-charging LIB. (a) Schematic illustration of the discharge state with graphite as the negative electrode and  $\text{LiCoO}_2$  as the positive electrode. (b) When a force is applied, the piezoelectric PVDF separator layer creates a piezo potential, with a positive charge at the positive electrode side and a negative charge at the negative electrode. (c) Under the driving of the piezoelectric field, the  $\text{Li}^+$  ions migrate from the positive electrode to the negative electrode, leading to the corresponding charging reactions at the two electrodes. (d) When the applied force is released, the piezoelectric field of the PVDF disappears, forming a potential difference between the oxidized  $\text{LiCoO}_2$  and the reduced graphite. (e) The movement of the external circuit electrons drives the lithium ions back to their original state and completes a discharge.

generation of an electric current, thereby establishing the foundation for self-charging technologies. Subsequently, various piezoelectric nanomaterials, including  $\text{GaN}$ ,  $\text{BaTiO}_3$ , and poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), were applied to fabricate PENGs [53–64]. The objective is to achieve energy self-sufficiency in electronic systems, where energy can be harvested to compensate for energy consumption by self-charging. This involves using a piezoelectric film as a separator in a capacitor or battery. The piezoelectric potential generated under pressure drives the ion movement in the electrolyte and initiates the electrochemical reaction in the electrode. This process realizes the conversion and storage of energy (electrical  $\rightarrow$  chemical), completing self-charging [65–70].

## 2.1. Piezoelectric nanogenerator (PENG)-based LIBs

In 2012, Professor Wang achieved a significant breakthrough by integrating PENGs and LIBs for the first time [71]. This pioneering work utilized a piezoelectric polyvinylidene fluoride (PVDF) polymer film as a separator, where the piezoelectric potential generated under mechanical strain drove the migration of lithium ions, enabling the charging process. Although this marked a critical advancement in PENG-based LIBs, the energy conversion efficiency from mechanical to electrochemical energy remained relatively low. Subsequent research has focused on improving energy conversion efficiency through improvements in device structure and piezoelectric separator design [72,73]. One strategy involves replacing the traditional stainless-steel casing with a flexible alternative that works efficiently at lower frequencies and pressures. For instance, Xue et al. developed a flexible self-charging LIB using a Kapton casing, which can harness energy from subtle human body movements, thereby minimizing mechanical energy loss [72]. Recent advancements have demonstrated significant progress, with the self-charging voltage increasing from 70 mV to 240 mV within 330 s under periodic tapping (6 N, 1 Hz) in a flexible configuration (6 N, 1 Hz) in a flexible case [73].

Another approach focuses on enhancing the specific surface area and piezoelectric sensitivity of piezoelectric films [74–79]. Two key strategies have been identified: 1) Designing Porous/Mesoporous PVDF Piezo-Separators: By incorporating geometric strain confinement effects and large surface area structures, these separators reduce charge-transfer resistance and accelerate  $\text{Li}^+$  ion transport, thereby improving energy conversion efficiency. 2) Incorporating Nano-Piezoceramics: The addition of high-performance piezoceramics, such as  $\text{PbZrO}_3$ ,  $\text{BaTiO}_3$ , and  $\text{BaZrO}_3$ , into PVDF films not only enhances the piezoelectric voltage but also facilitates the fabrication of porous structures, further optimizing performance.

The working mechanism of LIBs based on self-charging PENG is shown in Fig. 2. These batteries share a similar structure with conventional LIBs, except that the piezoelectric potential generated by deformation drives the migration of lithium ions through the piezoelectric separator. Taking the  $\text{LiCoO}_2$ /PVDF/graphite PENG-based LIB as an example, the initial state of the device is in a discharged condition (Fig. 2a). When the compressive stress is applied to the device, the PVDF piezoelectric separator layer generates a piezoelectric field, producing a positive piezoelectric potential on the  $\text{LiCoO}_2$  side and a negative piezoelectric potential on the graphite side (Fig. 2b). Under the influence of the piezoelectric field,  $\text{Li}^+$  ions migrate from the positive electrode to the negative electrode through the PVDF film separator within the electrolyte. This ion migration drives the reactions at the positive electrode ( $\text{LiCoO}_2 \rightarrow \text{Li}_{1-x}\text{CoO}_2 + x\text{Li}^+ + xe^-$ ) and negative electrode ( $6\text{C} + x\text{Li}^+ + xe^- \rightarrow \text{Li}_x\text{C}_6$ ). The  $\text{Li}^+$  migration ends until the piezoelectric field in PVDF is balanced by the  $\text{Li}^+$  ion distribution (Fig. 2c). Upon the removal of external pressure, a portion of the  $\text{Li}^+$  ions migrate back, the piezoelectric field in the PVDF gradually dissipates, and the self-charging process is completed. This results in a small charging voltage between the oxidized  $\text{LiCoO}_2$  and the reduced graphite (Fig. 2d). Next, the circuit is connected to complete the discharge process. In other words, the movement of the external circuit electrons drives the lithium

ions back to their original state (Fig. 2e).

The self-charging mechanism can be further elucidated from a thermodynamic perspective using the Nernst equation [75], the relationship between the electrode potential and the concentration of  $\text{Li}^+$  is expressed as follows,

$$\varphi(\text{Li}_{1-x}\text{CoO}_2/\text{LiCoO}_2) = \varphi^0(\text{Li}_{1-x}\text{CoO}_2/\text{LiCoO}_2) - \frac{RT}{F} \ln \frac{1}{a_{\text{positive electrode}}(\text{Li}^+)^x} \quad (1)$$

$$\varphi(\text{C}/\text{Li}_x\text{C}_6) = \varphi^0(\text{C}/\text{Li}_x\text{C}_6) - \frac{RT}{F} \ln \frac{1}{a_{\text{negative electrode}}(\text{Li}^+)^x} \quad (2)$$

Here,  $\varphi(\text{Li}_{1-x}\text{CoO}_2/\text{LiCoO}_2)$  and  $\varphi(\text{C}/\text{Li}_x\text{C}_6)$  represent the actual potentials, while  $\varphi^0(\text{Li}_{1-x}\text{CoO}_2/\text{LiCoO}_2)$  and  $\varphi^0(\text{C}/\text{Li}_x\text{C}_6)$  denote the standard potentials.  $R$  is the gas constant,  $T$  is the temperature, and  $F$  is the Faraday constant. The terms  $a_{\text{positive electrode}}(\text{Li}^+)$  and  $a_{\text{negative electrode}}(\text{Li}^+)$  represent the activities of  $\text{Li}^+$  around the positive and negative electrodes, which can be approximately equal to the concentrations. Driven by the piezoelectric field,  $\text{Li}^+$  ions migrate from the positive to the negative electrode. This migration leads to a decrease in the concentration of  $\text{Li}^+$  ions at the positive electrode and an increase at the negative electrode. As a result, the variation in the potential of the negative electrode,  $\varphi(\text{C}/\text{Li}_x\text{C}_6)$ , is greater than that of the positive electrode,  $\varphi(\text{Li}_{1-x}\text{CoO}_2/\text{LiCoO}_2)$ . This differential change in electrode potentials generates a self-charging voltage, enabling the energy storage.

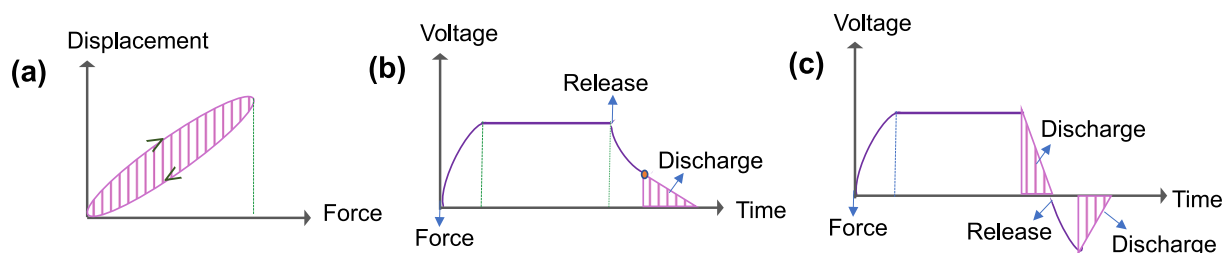
A critical question arises: Can the piezoelectric potential generated by PVDF effectively drive the intercalation and deintercalation of  $\text{Li}^+$  ions in  $\text{LiCoO}_2$  and graphite? This can be investigated through density functional theory calculations [77]. These simulations can model the piezoelectric field and its influence on the intercalation/deintercalation energies of  $\text{Li}^+$  ions. Studies have shown that the piezoelectric field can significantly reduce the energy barriers associated with  $\text{Li}^+$  ion intercalation and deintercalation, thereby facilitating these processes. These findings highlight the importance of optimizing the piezoelectric properties of materials like PVDF to enhance the efficiency of energy conversion and storage.

## 2.2. PENG-based capacitors

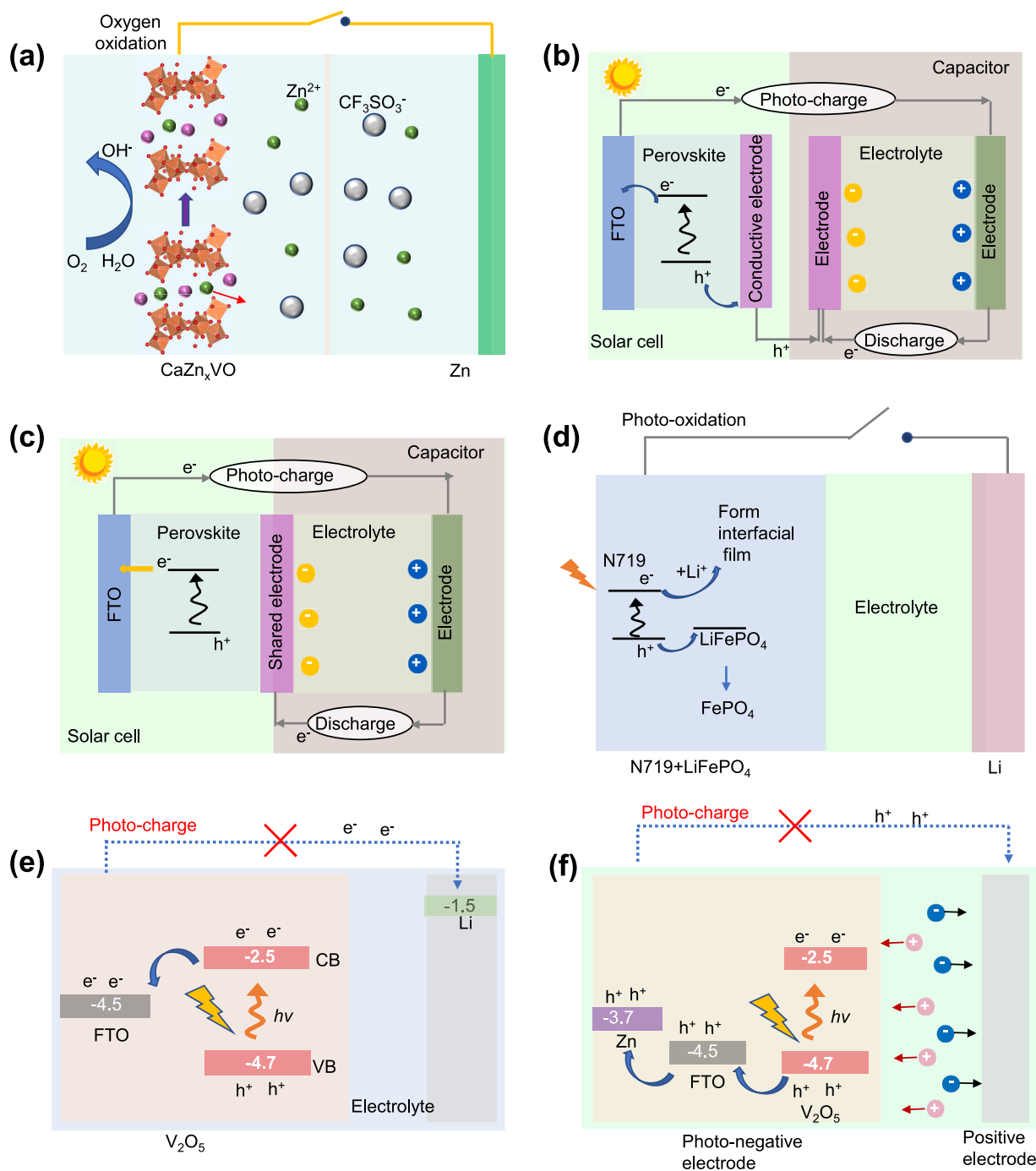
In PENG-based LIBs, energy conversion and storage are accomplished through the phase transition associated with  $\text{Li}^+$  ion intercalation and deintercalation in the electrode materials. This process typically requires a significant driving force to overcome the energy barriers involved. In contrast, capacitors, which store energy through

surface-based mechanisms such as adsorption–desorption or pseudocapacitive reactions, exhibit several distinct advantages [80,81]. 1) Lower reaction energies and faster charge transport, 2) Compatibility with environmentally friendly systems (aqueous solutions and hydrogels). This characteristic makes them suitable for applications in detection equipment related to the human body.

Similar to the piezoelectric separators used in PENG-based LIBs, separators in self-charging capacitors can also be constructed from polymeric piezoelectric films or gel electrolyte membranes embedded with piezoelectric ceramics. To enhance the piezoelectric effect of these separators and improve energy conversion efficiencies, several strategies have been employed. These include the fabrication of porous piezoelectric films, the nano-sizing of piezoelectric ceramics, and the development of composite materials combining piezoelectric polymers with piezoelectric ceramics [82–91]. The selection of electrode materials for these systems is highly diverse, encompassing carbon-based materials (e.g., activated carbon, carbon nanotubes, and reduced graphite), conductive polymers (e.g., poly(3,4-ethylene dioxithiophene) polystyrene sulfonate), and inorganic materials (e.g., metal oxides, nitrides, sulfides, phosphides, siloxene, and their composites) [92–96]. In 2015, the first self-charging symmetric capacitor based on a PENG was reported, using PVDF-ZnO piezoelectric separator and  $\text{MnO}_2$  electrodes. This system demonstrated the direct conversion of mechanical energy into electrochemical energy [82]. Subsequent studies have explored symmetric electrodes such as  $\text{Co-Fe}_2\text{O}_3$ ,  $\text{MoSe}_2$ ,  $\text{NiCo}_2\text{O}_4$ ,  $\text{MnO}_2$ , and siloxene in PENG-based symmetric capacitor [83–85,92,93]. However, electrode materials relying on intercalation/deintercalation mechanisms often suffer from slow reaction kinetics, requiring substantial driving forces and exhibiting low energy conversion efficiencies. To address these limitations, researchers have turned to alternative materials, such as carbon-based materials and conductive polymers, which exhibit rapid adsorption–desorption mechanisms. For instance, carbon nanotubes, activated carbon, and graphene electrodes enable fast ion adsorption and desorption, achieving energy conversion and storage rates faster than traditional Faradaic reactions and intercalation mechanisms [86–88]. Additionally, symmetric self-charging capacitors based on conductive polymers are under active development, with flexible polymer electrodes showing promise for integration into flexible electronic devices [95]. Asymmetric self-charging capacitors have been proposed [89–91] to meet the growing demand for high energy density. In these systems, one electrode typically employs carbon materials or conductive polymers with rapid adsorption–desorption capabilities, while the other electrode incorporates materials with high-



**Fig. 3.** The working mechanism of PVDF piezoelectric generator. (a) Hysteresis loop diagram of force versus displacement. (b,c) Variation curve of voltage across PVDF with time (Assuming that the piezo voltage is proportional to the force). b, Discharge after releasing the force. c, Discharge first, then release the force, and discharge again.



**Fig. 4.** Mechanism of chemical oxidation self-charging energy storage. (a) The working mechanism of chemically self-charging  $\text{CaVO//Zn}(\text{CF}_3\text{SO}_3)_2/\text{Zn}$  under neutral aqueous solution. (b) Connection of the solar cell and capacitor through an isolated design. (c) Connection of the solar cell and capacitor through a three-electrode system. (d) Photo-oxidation process of  $\text{N719/LiFePO}_4$  positive electrode in LIBs under sunlight. (e) Unreasonable photo-charging mechanism based on electron conduction. (f) Photo-charging mechanism based on hole conduction.

energy-density intercalation or pseudocapacitive mechanisms. This asymmetric design balances the need for high power density and high energy density, making it a promising approach for next-generation energy storage systems.

Although self-charging capacitors can be designed in symmetric and asymmetric configurations, their underlying mechanisms share fundamental similarities. Taking a symmetrical self-charging capacitor as an example, the discharge state is the initial state, with activated carbon electrodes separated by a PVDF piezoelectric film. The electrolyte, containing ions such as  $\text{Na}^+$  and  $\text{Cl}^-$ , is uniformly distributed within the system. When an external force is applied, the PVDF piezoelectric film

undergoes polarization, leading to the separation of positive and negative charges and the generation of a piezoelectric potential. This potential drives the migration of ions in the electrolyte: negative ions ( $\text{Cl}^-$ ) move toward the positive electrode, while positive ions ( $\text{Na}^+$ ) migrate toward the negative electrode, where they undergo adsorption reactions. Ion migration ceases until the driving force of the piezoelectric potential generated under pressure is insufficient. Upon removal of the external force, the piezoelectric potential dissipates, and the self-charging process is completed. When the circuit is connected, the potential difference between the electrodes drives the discharge process.

The self-charging mechanism can be explained through the Nernst



equation [85], which describes the relationship between the electrode potential difference and the ion concentration in the electrolyte.

In the normal state,

$$\varphi(\text{activated carbon}) = \varphi^0(\text{activated carbon}) - \frac{RT}{F} \ln \frac{X}{1-X} \quad (3)$$

When applied a force,

$$\varphi(\text{activated carbon}) = \varphi^0(\text{activated carbon}) - \frac{RT}{F} \ln \frac{X^0}{1-X^0} \quad (4)$$

Here,  $\varphi(\text{activated carbon})$  represents the actual potential of the two activated carbon electrodes, while  $\varphi^0(\text{activated carbon})$  represents the standard potential.  $R$ ,  $T$ , and  $F$  are gas constant, temperature, and Faraday constant, respectively.  $X$  represents the concentration of ions migrated to the positive and negative electrode surfaces, while  $X^0$  denotes the concentration after the force is applied. When a force is applied,  $X^0$  is greater than  $X$ , leading to an elevation in the potential of two activated carbon electrodes. However, due to differences in the adsorption behavior of  $\text{Na}^+$  and  $\text{Cl}^-$  ions at the positive and negative electrodes, a potential difference arises, resulting in the self-charging effect.

Compared to LIBs based on self-charging nanogenerators, capacitors demonstrate superior performance in terms of faster reaction kinetics, longer cycle life, and enhanced environmental compatibility. These advantages significantly broaden their application scope, particularly in flexible wearable electronic devices, especially when integrated with water-based gel polymer electrolytes. However, it is important to note that the working principle of PVDF-based piezoelectric generators relies on converting the net mechanical work—represented by the area enclosed within the “force-displacement” hysteresis curve of the elastic body (shaded red in Fig. 3a)—into other forms of energy, such as heat and electricity (Fig. 3). Assuming that the piezoelectric voltage is proportional to the applied force, the energy conversion process can be analyzed as follows: in the presence of an electrolyte, the discharge of the generator (shaded red in Fig. 3b) is minimal after the force is released. When the force is applied, the external circuit discharges to zero voltage, and upon removal of the force, the voltage becomes negative, triggering a second external discharge (Fig. 3c). The total energy generated from these two discharges is constrained by the area enclosed within the hysteresis loop in Fig. 3a. As a result, the energy conversion efficiency of such systems remains relatively low, typically ranging between 5–20 %. This low efficiency highlights a critical challenge in the design of piezoelectric energy harvesting systems. While capacitors offer advantages such as rapid energy delivery and compatibility with flexible electronics, the inherent limitations of piezoelectric materials—such as energy losses due to hysteresis and incomplete conversion of mechanical work into electrical energy—must be addressed to improve overall performance. Strategies to enhance energy conversion efficiency, such as optimizing the piezoelectric properties of materials, reducing hysteresis losses, and improving device architecture, are essential for advancing the practical applications of self-charging capacitors in wearable and flexible electronic devices.

### 3. Chemical oxidation self-charging energy storage

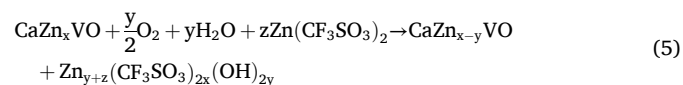
The zinc-air battery, first developed in 1878, is an important milestone in the participation of oxygen in energy storage. This type of battery utilizes oxygen from the air as the positive electrode material and metallic zinc as the negative electrode material, making it a unique and efficient chemical power source. Subsequently, other metal-air batteries, such as those on Li, Fe, and Al, were gradually developed [97]. Despite the successful utilization of oxygen in these systems, a major limitation remains: the need for external recharging. However, with the continuous advancement and diversification of electrode materials, certain materials have garnered significant attention due to their ability to undergo oxidation by atmospheric oxygen at low valence

states. This property enables a novel charging mechanism, where the battery can be recharged through direct oxidation by oxygen in the air, eliminating the need for external power sources [98–108].

#### 3.1. Oxygen oxidation

In 2014, the discovery of Prussian white spontaneously oxidizes to Prussian blue in the presence of dissolved oxygen in an aqueous solution, laying the foundation for a self-rechargeable battery [98]. Until 2020, a chemically self-charging aqueous zinc ion battery (ZIB) was designed using the  $\text{CaV}_6\text{O}_{16} \cdot 3\text{H}_2\text{O}$  ( $\text{CaVO}$ ) as the positive electrode material [99]. Vanadium-based compounds were selected due to their open-framework crystal structures, multiple oxidation states of vanadium, and their sensitivity to oxidation in the low-valence state. The conversion and storage of chemical energy to electrical energy are realized by directly exposing the positive electrode material ( $\text{CaVO}$ ) to air for an auto-oxidation reaction. Following this, other vanadium-based materials, including  $\text{V}_6\text{O}_{13}$ , and  $\text{VO}_2$ ,  $\text{V}_2\text{O}_5$ , were also explored for self-charging energy storage in ZIBs or zinc-ion capacitors [100–102].

With the material system identified, our primary focus has shifted to understanding the underlying mechanism of oxidative self-charging, using  $\text{CaVO}$  as an example among vanadium-based compounds. In redox reactions, electron transfer is driven by the redox potential difference ( $\Delta E$ ) between reactants. Cyclic voltammetry (CV) curves reveal that the minimum reduction peak potential of  $\text{CaVO}$  with  $\text{Zn}^{2+}/\text{Zn}$  is 0.47 V. The redox potential of  $\text{CaZn}_x\text{VO}$  is calculated to be  $-0.27$  V versus the standard hydrogen electrode (SHE) using the Nernst equation [99]. When the potential of the oxidant exceeds  $-0.27$  V,  $\text{CaZn}_x\text{VO}$  undergoes spontaneous oxidation. The standard electrode potentials of  $\text{O}_2$  in neutral and acidic media are  $\sim 0.4$  V and  $\sim 1.23$  V versus SHE, which are higher than that of  $\text{CaZn}_x\text{VO}$ . This indicates that  $\text{CaZn}_x\text{VO}$  tends to release electrons for oxidation, while  $\text{O}_2$  can accept these electrons for reduction. The self-charging reaction equation in a neutral environment can be described as follows (Fig. 4a):



In this reaction,  $\text{O}_2$  accepts the electrons from  $\text{CaZn}_x\text{VO}$ , leading to the disinsertion of zinc ions. Simultaneously,  $\text{O}_2$  reacts with the electrolyte to generate  $\text{OH}^-$  ions, which combine with the extracted zinc ions and other electrolyte components to form an amorphous interface film composed of  $\text{Zn}_{y+z}(\text{CF}_3\text{SO}_3)_{2x}(\text{OH})_{2y}$ . In acidic environments, the mechanism differs slightly, as oxygen is reduced to  $\text{H}_2\text{O}$  in the presence of  $\text{H}^+$  ions.

To enhance the self-charging efficiency, researchers have focused on materials with fast adsorption-desorption mechanisms, such as conducting polymers and carbon-based materials. Nitrogen-containing conducting polymers are commonly used in self-charging systems. Taking C=N-containing polymers as an example, C-N- $\text{M}^{2+}$  is formed after discharge (where M ions depend on the electrolyte composition) [103]. Under open-circuit voltage (OCV) state, exposure to  $\text{O}_2$  causes C-N- $\text{M}^{2+}$  to revert to C=N, forming MO on the polymer surface. This process increases the electrode potential, realizing the chemical oxidation self-charging process. Similarly, in carbon-based material,  $\text{Zn}^{2+}$  ions are adsorbed onto the carbon surface during discharge (C// $\text{Zn}^{2+}$ ) [104]. When exposed to air, an oxidation reaction occurs, consuming electrons and removing  $\text{Zn}^{2+}$  ions, resulting in the formation of zinc oxide on the electrode surface as part of the solid electrolyte interphase film. In addition to material design improvements, another common strategy to enhance self-charging efficiency involves increasing the concentration of oxygen and electrolytes [105–107]. By optimizing these parameters, the electrochemical reactions driving self-charging can be significantly accelerated. Furthermore, future advancements in self-charging systems may be achieved by combining innovative material designs with additives that enhance oxygen fixation, thereby

improving energy conversion efficiency [109].

### 3.2. Photo-oxidation

Introducing sunlight into the system introduces complexity, often leading to misconceptions among readers. It is therefore essential to provide a clear and accurate understanding of the underlying principles. The integration of solar cells with energy storage batteries enables a multi-step energy conversion and storage process (solar → electrical → chemical → electrical). This concept has been achieved through two design approaches: an isolated design and a three-electrode system [110]. In the isolated design, photovoltaic (PV) solar cells and the electrochemical (EC) batteries operate as independent units, interconnected via external wires. In contrast, the three-electrode system integrates the two components through a shared electrode. This shared electrode serves a dual function: facilitating photoelectric energy conversion in the PV component and enabling electrochemical energy storage in the EC component. Consequently, the properties of the shared electrode—such as electrical conductivity, energy band alignment, and chemical compatibility—play a critical role in determining the overall energy conversion and storage efficiency of the system.

The mechanism of the isolated PV-EC design is shown in Fig. 4b, where electron-hole separation occurs in the photosensitive layer of the solar cell part under sunlight [110]. Electrons are moved to the negative electrode and the holes migrate to the positive electrode through the external circuit. The negative electrode undergoes reduction, and the positive electrode is oxidized to complete the charging process. When the energy storage device is turned on, the stored electrochemical energy is converted into electrical energy through discharge. This system accommodates a wide range of photosensitive materials, including perovskite, crystalline silicon, copper indium gallium selenide, organic photovoltaic materials, and dye-sensitized materials. For the EC module to be effectively charged by the PV module, the open-circuit voltage (OCV) of the PV component must exceed the electromotive force of the EC module. This requirement often necessitates the use of tandem photovoltaic cells to achieve the necessary voltage levels. This concept also applies to the three-electrode system (Fig. 4c).

Recently, a two-electrode system has been proposed, in which one electrode employs an integrated or hybrid material for photosensitive and energy storage [111–123]. It is important to note that there are two necessary conditions for photo-charging: 1) A conductive pathway must exist between the positive and negative electrodes during the photo-charging process, and 2) The energy levels of the positive and negative electrodes, including their conduction band and valence states, must match. In an open circuit between the electrodes, a photo-oxidative mechanism happens [111,123]. As shown in Fig. 4d, liquid LIBs were assembled using a mixture of N719 dye and  $\text{LiFePO}_4$  as the positive electrode and Li metal as the negative electrode [111]. When illuminated under OCV, N719 produces electron-hole separation. The holes drive the delithiation of  $\text{LiFePO}_4$ , and electrons participate in the formation of the interfacial film on the electrode surface. At this point, the positive electrode completes the charging process through photo-oxidation. Upon connecting the positive and negative electrodes, the conversion and storage of solar energy can be realized.

Currently, research efforts are focused on metal-based batteries utilizing Li and Zn as negative electrodes. Some studies propose a mechanism in which photogenerated electrons migrate to the metal negative electrode through an external circuit, driving internal ions ( $\text{Li}^+$ ,  $\text{Zn}^{2+}$ ) to migrate to the negative electrode and complete the photo-charging process. However, this mechanism faces a fundamental challenge: electrons naturally tend to move toward lower energy states. For instance, it is thermodynamically unfavorable for electrons from conductive glass (e.g., fluorine-doped tin oxide, FTO, with a work function of approximately  $-4.5$  eV) to migrate to Li ( $-2.9$  eV) or Zn ( $-4.3$  eV) (Fig. 4e) [112–119]. A related question is whether solar energy conversion and storage can be effectively realized in capacitor-

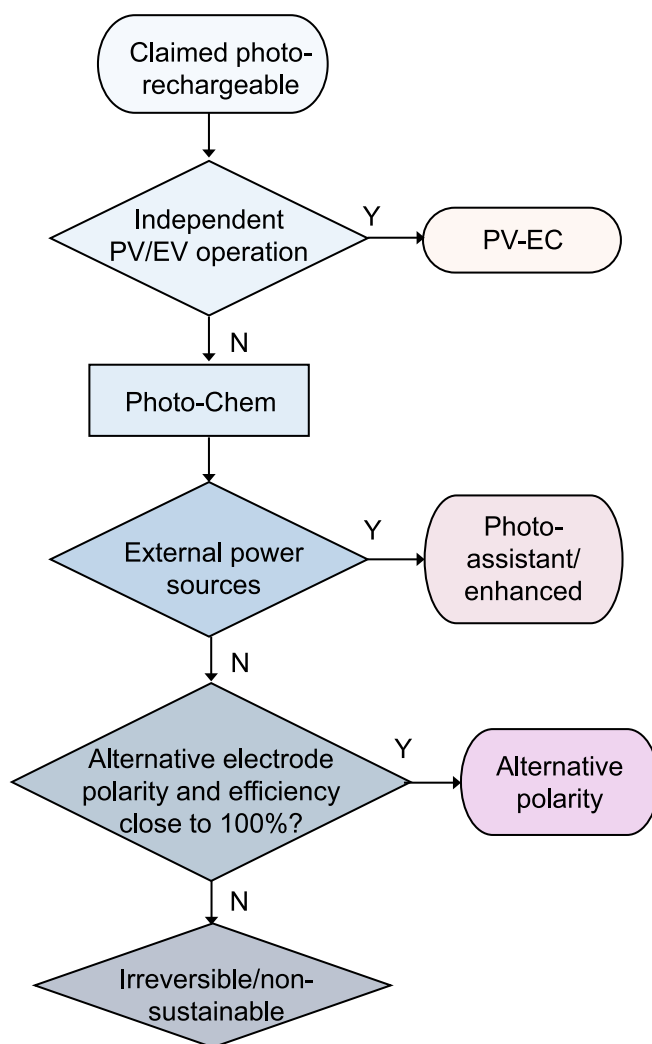
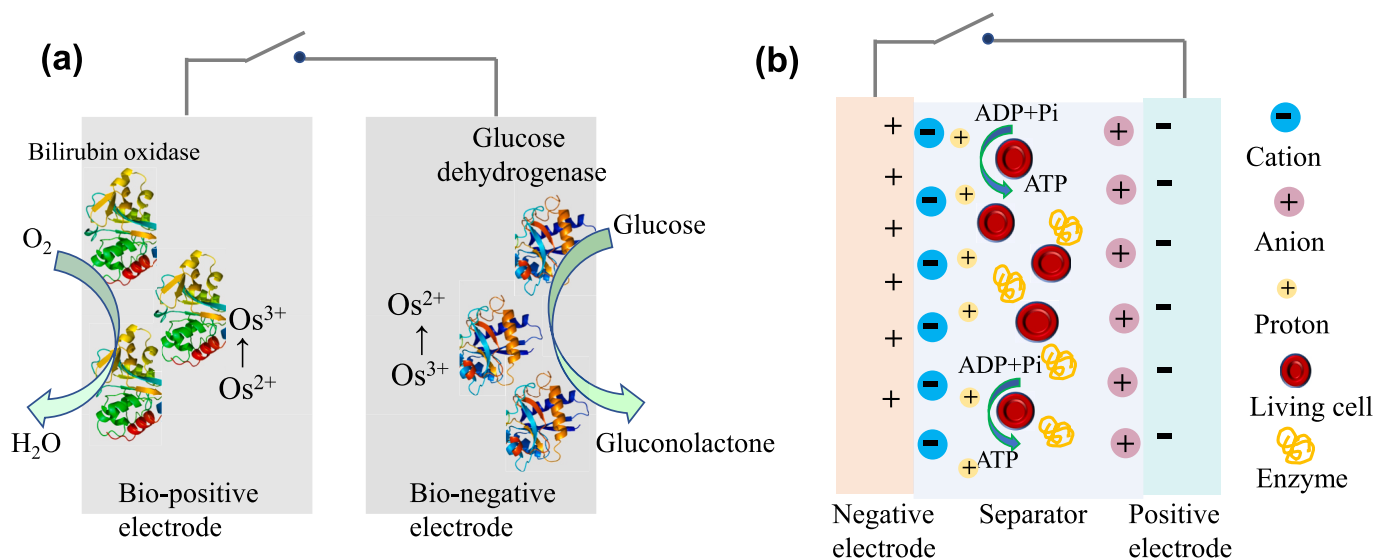


Fig. 5. Suggested protocol for classification of photo-rechargeable batteries.

based systems [120–122]. As early as 2004, Murakami proposed the photo-capacitors [120], but the underlying mechanism was not elucidated at the time. Later, Volder's group attempted to explain the photo-charging mechanism in capacitors, but their explanation appears inconsistent with established physical principles [121]. They suggested that the migration of holes migrates from low-energy bands to high-energy bands, proposing that photogenerated holes from the photo-negative electrode could migrate to the positive electrode via the external circuit under illumination, driving the adsorption of negative ions from the electrolyte onto the positive electrode surface. Simultaneously, the remaining electrons on the photo-negative electrode would attract positive ions to complete the photo-charging process (Fig. 4f). However, this explanation is problematic because photogenerated electrons cannot spontaneously migrate to the negative electrode, nor can photogenerated holes move to the positive electrode without an external driving force. These inconsistencies highlight the need for caution when interpreting proposed mechanisms in the literature, as some explanations may lack scientific rigor. It is essential to ensure self-charging photo-response batteries develop on a healthy and scientific track.

Compared to energy storage batteries, energy storage capacitors offer several advantages, including miniaturization, enhanced safety, non-toxicity, and a longer service life, enabling them to operate continuously around the clock. Integrated self-charging capacitors based on chemical oxidation and photo-oxidation mechanism hold great



**Fig. 6.** The working mechanism of self-charging nanocapacitors catalyzed by biological enzymes. (a) Schematic representation of poly(vinyl imidazole-co-allylamine)[Os(bpy)<sub>2</sub>Cl]-based Nernstian BSC during charging. (b) Schematic illustration of ion transport and energy storage mechanism between two working electrodes in blood.

potential for applications in outdoor detection devices, including infrared detectors, outdoor cameras, and laser weapons. However, it is crucial to recognize that self-charging behavior—whether driven by oxidation or photo-oxidation mechanisms—under open-circuit voltage (OCV) conditions can lead to the continuous consumption of the negative electrode or electrolyte. Once the negative electrode or active ions in the electrolyte are depleted, the reaction ceases entirely. To achieve long cycle life, it is necessary to incorporate periodic electric current charging after a certain number of cycles or to enhance the reversibility of oxidation products. However, complete reversibility is inherently unattainable due to the fundamental principle of energy conservation. Notably, some published studies, often titled “photo-rechargeable,” claim to eliminate the need for an external power source and appear to demonstrate recyclability. However, in the absence of reverse current, these systems are neither sustainable nor truly recyclable, effectively representing a misleading interpretation of their functionality. To address these challenges, a protocol for rapidly assessing the validity of such photo-rechargeable systems is proposed (Fig. 5). This protocol emphasizes the necessity of chemical and current reversibility as key criteria for evaluating the feasibility and sustainability of these systems. By adhering to such rigorous standards, the field can advance toward more reliable and scientifically sound energy storage solutions.

#### 4. Self-charging nanocapacitors catalyzed by biological enzymes

Health detection in the human body, such as heart rate, blood pressure, and blood sugar, has become an important research direction. For these sensors to be effective, they must be miniaturized to the nanoscale to avoid disrupting biological functions. This miniaturization necessitates equally compact power sources that do not rely on external charging. Given the human body’s saline-like environment, water-based capacitors are preferable over organic-based batteries due to their better safety and biocompatibility. Therefore, the development of self-charging biocompatible batteries or nanocapacitors is imminent [124–130]. The design of capacitor electrodes for such applications must satisfy two conditions: 1) Biocompatibility, and 2) the ability to undergo spontaneous redox reactions in the presence of biological enzymes. A significant breakthrough came in 2014 when the Cosnier group developed a supercapacitor using wired enzymes on carbon nanotube substrates, achieving a self-charging voltage of up to 1 V. This highlighted the feasibility of biological self-charging capacitors [131]. However, due to

concerns over the slight biotoxicity of carbon nanotubes, subsequent research has shifted toward biocompatible polymer materials for capacitor electrodes. Depending on the required energy density, these electrodes can be configured as either symmetrical or asymmetric polymer electrodes [132–134]. Moving closer to practical applications, Schmidt and colleagues designed a tubular nano-bio supercapacitors in 2020 [135]. The energy storage in nano-bio supercapacitors occurs between two thin flexible poly(3,4-methylenedioxy-thiophene)-poly(styrene sulfonate) redox electrode layers, using photopatterned poly(vinyl alcohol) as proton exchange separators and blood as the working electrolyte. It can deliver up to 1.6 V in blood, powering a complex integrated sensor system for measuring pH in the blood.

The intrinsic reaction mechanism can be elucidated through two typical examples. As shown in Fig. 6a, a biodevice utilizing poly(vinyl imidazole-co-allylamine) as bio-positive and bio-negative electrodes, along with two different biocatalysts, achieves self-charging by establishing an activity gradient of redox polymer-bound Os complexes. Specifically, at the bio-negative electrode, glucose oxidation is catalyzed by glucose dehydrogenase, leading to the reduction of Os ( $\text{Os}^{3+} \rightarrow \text{Os}^{2+}$ ). Simultaneously, at the bio-positive electrode,  $\text{O}_2$  is reduced to  $\text{H}_2\text{O}$  by bilirubin oxidase, resulting in Os oxidation ( $\text{Os}^{2+} \rightarrow \text{Os}^{3+}$ ). This redox activity creates a potential difference between the two bioelectrodes, completing the self-charging process. To prevent self-discharge when integrated into the blood system, an ion exchange membrane is incorporated. In Fig. 6b, the self-charging mechanism of biological supercapacitors is driven by enzyme-mediated redox reactions in the blood. These reactions induce the migration of positive charges toward the bio-positive electrode and negative charges toward the bio-negative electrode, thereby completing the self-charging process.

Inspired by the advancements in bio-nanocapacitors, self-powered energy systems offer promising solutions to several challenges in the biological field. These include: 1) Detection of Biomarkers: Micro-detectors powered by such systems can monitor glucose, uric acid, and lactic acid levels in the blood, enabling early detection of diseases or cancers. 2) Self-Powered Medical Devices: Miniature devices, such as cardiovascular stents, can operate autonomously to support normal physiological functions. 3) Intelligent Micro-Robots: These devices can monitor human physiological information and transmit data externally, facilitating real-time health monitoring. However, challenges remain. Enzyme catalysis, a key component of these systems, is not entirely reversible, and enzyme activity must be carefully regulated to ensure



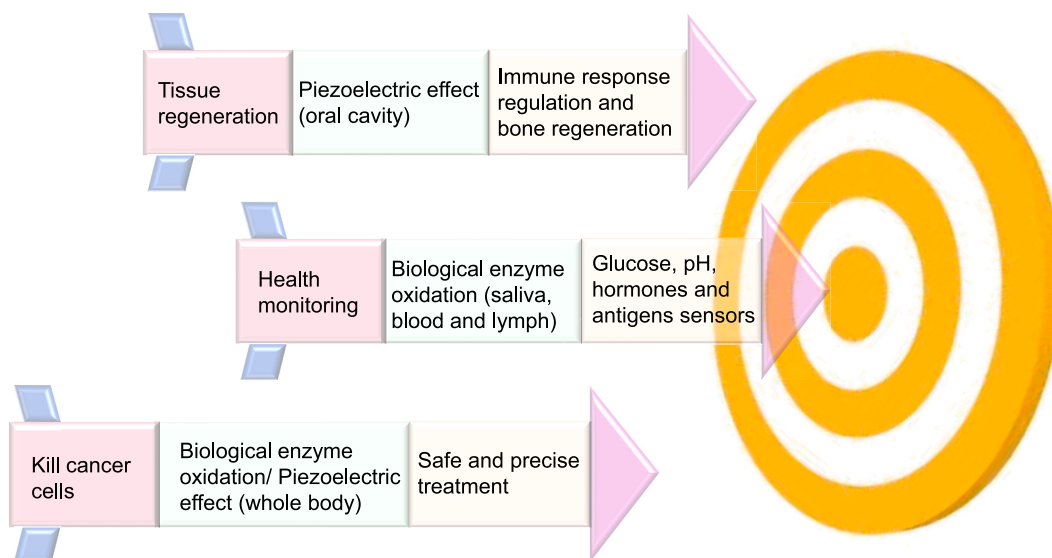


Fig. 7. Future applications of self-charging energy storage devices.

optimal performance. Next, the design of biodegradable polymer materials is critical to ensure that these devices do not cause harm to the body after their service life is complete. In addition, due to capacity and voltage limitations, the device structure needs to be optimized.

## 5. Summary and outlooks

This concise review provides a profound introduction to integrated self-charging batteries and capacitors, exploring materials selection for their working mechanism and providing insights into various self-charging technologies. It also critically evaluates and addresses certain unsubstantiated claims found in published literature. The review highlights three primary mechanisms: physical piezoelectricity, chemical oxidation, and biological enzyme catalysis. Physical piezoelectricity self-charging batteries and capacitors are designed using materials based on the piezoelectric effect. In this process, the piezoelectric effect membrane drives electrolyte ions toward the electrode, initiating redox reactions. Chemical oxidative self-charging batteries and capacitors employ materials that can undergo spontaneous oxidation. The low-valence discharge products are spontaneously oxidized to high-valence states by oxygen in the air, completing the self-charging process. The introduction of photosensitive materials enables photo-oxidative self-charge and energy storage. Under illumination conditions, electron-hole separation occurs in the photosensitive material. The holes contribute to the oxidation of the electrode material, while electrons participate in forming an interfacial film on the electrode surface. However, it is important to note that current challenges exist with the mechanism of two-electrode photo-chargeable batteries in closed circuits. Biological self-charging capacitors based on biocompatible polymer electrodes are driven by redox-active enzymes in living organisms. This review not only provides a solid theoretical foundation for the development of self-charging energy storage devices but also outlines future directions for the field (Fig. 7).

(1) *Health monitoring.* Self-charging nanocapacitors are assembled using biocompatible polymers as electrode materials, and body fluids such as electrolyte solutions (saliva, blood, and lymph) along with biocompatible hydrogels as separators. In an OCV state, redox enzymes in bodily fluids initiate biological reactions that drive the movement of positive and negative charges in bodily fluids toward the bio-positive

electrode and bio-negative electrode. This results in an increase in the potential difference between the two electrodes, enabling a self-charging process that powers the sensor to detect glucose, pH, hormone, and antigen levels in bodily fluids. The purpose is to detect potential diseases and prevent their occurrence.

(2) *Tissue regeneration.* To address oral issues such as poor alveolar bone shape or bone defects, a self-charging capacitor leveraging the piezoelectric effect can be devised to generate electrical signals facilitating the regulation of immune responses such as guidance of macrophage polarization and promoting bone regeneration. This design employs biocompatible polymers as electrode materials, PVDF piezoelectric membranes as diaphragms, and saliva as electrolytes. The pressure from tooth chewing induces the piezoelectric effect in the PVDF membrane, separating charges and causing positive and negative charges in the saliva to adsorb onto the electrode material, thereby achieving self-charging. The discharge process involves the output of electrical signals, which contributes to the regulation of immune microenvironment and promotion of tissue regeneration.

(3) *Cancer treatment.* Cancer represents a critical challenge in contemporary medicine, and the precise eradication of cancer cells is an urgent issue to address. In this context, self-charging energy storage devices implanted in the human body can emerge as a significant solution. Specifically, self-charging nano supercapacitors leveraging the piezoelectric effect or biological enzyme catalysis can be designed and implanted in vivo tailored to specific cancer sites. The voltage generated through self-charging is then converted into heat, enabling the precise and localized destruction of cancer cells. This approach, in comparison to conventional radiation therapy and drug treatments, offers a more body-friendly and targeted intervention.

We anticipate significant breakthroughs in the biological field through the integration of water-based self-charging capacitors. While self-charging energy storage devices are pivotal for the aforementioned applications, their practical implementation is inherently dependent on advancements in other microelectronic components and control chips. The synergy between these technologies is essential for realizing the full potential of self-charging systems in real-world applications.

## CRediT authorship contribution statement

**Jun Pan:** Writing – original draft, Project administration, Methodology. **Kaidi Yuan:** Methodology. **Ningjuan Ouyang:** Methodology. **Shixue Dou:** Writing – review & editing, Methodology. **Nana Wang:** Writing – review & editing, Project administration, Methodology, Conceptualization. **Srinivasan Madhavi:** Writing – review & editing, Project administration, Methodology, Conceptualization.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Data availability

No data was used for the research described in the article.

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