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Customizable Electrochemical Energy Storage Devices

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ABSTRACT: Customizable electrochemical energy storage device is a key component for the realization of next-generation wearable and bio-integrated electronics. This perspective begins with a brief introduction of the drive for customizable electrochemical energy storage devices. The following section traces the first-decade development trajectory of the customizable electrochemical energy storage devices. It then discusses the challenges and the future directions, calling for such devices that allow users to select, design and change the properties (including capacity, flexibility, shapes, and functionalities) according to real-life needs. Leveraging these customizable electrochemical energy storage devices will shed light on smarter programmable electrochemical energy storage devices to power future wearable and bio-integrated electronics.

The drive for customizable electrochemical energy storage devices

With technological progress and increasingly human-oriented advancements, electronics have transformed from being immobile and bulky to being portable and compact, becoming user-friendly and easy to operate. It underlines the ongoing emergence of wearable and bio-integrated platforms, with an emphasis to guide the current electronics to a higher degree of customizability in terms of flexibility, shape, and bio-conformability (**Figure 1**). To intimately integrate with the human body, electronics are becoming flexible and stretchable to withstand the large strain of deformation during complex body motions, allowing users to bend, twist and stretch the electronics without compromising their functions. Apart from flexibility and stretchability to comply with body motion, the human-oriented electronics also gear towards having customizable shapes for optimized space-utilization for different needs, such as near-body, on-skin, and implantable applications. Moreover, the customizable bio-conformability guarantees the electronics the desired bio-interface with human body, where it needs to be comfortable, biocompatible, implantable or even ingestible in some applications. Success in such customizability of electronics offers exciting opportunities to fabricate wearable electronics, epidermal electronics, implantable electronics and ingestible electronics for personalized fitness and biomedical applications (e.g., activity tracking, health monitoring, and controlled drug delivery).¹⁻¹⁰

As more wearable and bio-integrated electronics are expected to be manufactured and used, there is an increased demand for complementary electrochemical energy storage (EES) devices, especially batteries and supercapacitors. As a result of the evolution of customizable electronics, the customizability in flexibility, shape, and bio-conformability with human body has emerged as a new figure-of-merit for the future EES devices. The customizable EES devices are stretching the concept beyond capacity and rate capability, to an extent where they are adaptable with wearable

and bio-integrated electronics. Such customizable EES devices should connect seamlessly with customizable electronics in order to obtain truly customizable integrated systems for human body application. They present a new alternative in how the electronics are charged and operated (Figure 1).¹¹⁻¹³ To explore the future of customizable EES devices, it is worth revisiting the journey thus far, before we map out the path forward.

The first-decade journey towards customizable EES devices

It has been almost one decade since the first development of customizable EES devices. In the past decade, researchers have turned to endow EES devices with new form factors, making it flexible, malleable and biocompatible to be worn or bio-integrated (Figure 2). Initially, the development of customizable EES devices was mainly focused on improving the mechanical flexibility of devices (Figure 2). In 2007, a fully flexible EES device was developed by Pushparaj et al., wherein the nanocellulose paper embedded with aligned carbon nanotube (CNT) electrode and electrolytes offers mechanical bendability of the EES device during operation.¹⁴ A major step forward for customizable EES devices is to advance from mechanically flexible to stretchable, allowing harsher mechanical stimulation such as twisting and stretching. It was not until 2009 that the first stretchable EES device was demonstrated, partially because of the great challenges in selecting mechanically elastic active electrode materials for stretchable electrodes.¹⁵ Even using novel materials such as CNT and graphene, the maximum elastic strains possessed by the key active materials (e.g., <10% for CNT and <5% for graphene) in electrodes of supercapacitors and batteries still fall short of accommodating the substantial deformation (much greater than 100%) required by stretchable EES devices.¹⁶ To bridge the gap, researchers exploited two effective and conceptually different but mutually complementary methods to construct stretchable EES devices. One utilizes advanced nanomaterials, such as CNTs, graphene, other low-dimensional

nanostructured materials (e.g., metal nanowire) and their composites, to combine with intrinsically elastic polymers for improving electrochemical and mechanical properties of electrodes and electrolytes. The other focuses on devising buckling structure configurations comprising pre-stretched wave-like structures and bridge-island structures to impart stretchability to the brittle energy storage systems.^{12, 17} The arrival of these novel nanomaterials and rational structural designs gives EES devices a new lease of life and bring to a reality the stretchable variants.

Apart from the mechanical flexibility and stretchability, the conventional planar EES devices¹⁸ witnessed an evolution in their shape: Researchers focused on fabricating EES devices with shapes geometrically similar to the common fabric and wearable accessories. One trend is to miniaturize EES devices to dimensions of tens to hundreds of micrometers, which allows them to be packed into a microscale space and inconspicuously integrated with wearable electronics.¹⁹ Another trend in customizable EES devices is to embed them into textiles. By dyeing fabric sheets with CNT ink through a simple dip-and-dry process, highly conductive textiles as a platform for wearable supercapacitor were first demonstrated.²⁰ The textile structure also provided the textile EES devices with a stretchability of up to 120%. Generally, there are four levels (i.e., fibers, yarns, fabrics, and textile products) of hierarchical structures in textiles. Even though the electrochemical capacity and mechanical stretchability can be engineered into textiles at one or more of the hierarchical levels, the most effective and unobtrusive method remains engineering electrochemical functionalities at the fiber level without losing the accompanying desired attributes such as texture and comfort. EES devices assembled at fiber level perfectly inherited the desired attributes of textile (such as flexibility, stretchability, breathability, and comfort) while the electrochemical performance was not compromised. Hence, instead of endowing EES devices with mechanical resilience at the textile level, designing fiber-shaped supercapacitors represents a

bottom-up approach towards customizable EES devices.²¹ To further augment the inherent stretchability of the fiber-shaped EES devices, coiled spiral fiber-shaped electrodes can be employed, giving rise to spring-like elasticity. Furthermore, by combining the fiber-shaped electrodes with buckling structure and ultra-stretchable polymers, the stretchability of the customizable EES devices is dramatically increased. The shape of these devices also moved from fiber (one-dimensional, 1D) and textile (two-dimensional, 2D) into three-dimensional (3D) for better spatial utilization. Inspired by the art of folding paper into 3D structures, the paper-folding (origami) technique has been used to fabricate compact customizable EES devices with increased energy per footprint area and stretchability, representing the fusion of foldability and 3D shapes.²²⁻
²⁴ Similarly, paper-cutting or kirigami is further proposed to fabricate stretchable and tailorable EES devices.²⁵⁻²⁷ Apart from paper craft to design 3D EES devices, researchers continued to explore the fully 3D EES devices based on 3D CNT coated cellulose nanofibril aerogels²⁸ and polyaniline-CNT-sponge electrodes²⁹. The porous 3D electrode structures enable the EES devices with both arbitrary 3D shapes and compressibility. Bioinspired, spine-like shape and expandable honeycomb structures can further enable the 3D-shaped EES devices towards being more flexible and stretchable.³⁰⁻³¹

The evolution in the flexibility and shape of the customizable EES devices marked a significant scientific endeavor in the first decade. The marriage of EES devices with bio-conformable properties (biodegradable³², breathable³³, washable³⁴, aesthetic³⁵ and biocompatible³⁶⁻³⁷) presents another significant milestone towards a higher degree of customizability. Such advancements demonstrated EES devices to be worn on the wrist or head near the body, pasted on the skin,³⁶ implanted inside the human body,³⁸ or even ingested by individuals.³⁹⁻⁴⁰ The first-decade story of the wearable EES device is one of convergence of

customizability. Advances in materials, electrode structures, device fabrication and system integration each created the technology building blocks that subsequently enhanced the present customizable EES devices. We are now at a critical point where the design of EES device needs to be intertwined with real-world applications for creating a user-customizable device. Recently, an editable supercapacitor is developed to retain its functionality after being folded, cut and stretched.¹² The editable EES devices with device-level programmability, unlike the currently predesigned devices, allow wearers to remodel its shape and structure/stretchability to be compatible with various wearable electronics, indicating the convergence of the customizable properties (bendable, twistable, foldable, tailorable, stretchable, fabric, aesthetic, 3D-shaped, etc.) for customizable EES devices. In the future, allowing users to select, design and change its functions according to wearable and bio-integrated scenarios represents the inevitable direction towards next-generation customizable EES devices. Based on current research progress, some of the existing problems and future directions for customizable EES devices are discussed as follows.

Innovations in materials and structures of customizable devices

The primary function of EES devices is to store and supply energy. Intensive ongoing research aims to bring new materials with even higher specific capacity for next-generation customizable EES devices.⁴¹ However, in reality, the energy storage performance of customizable EES devices are usually much poorer than conventional counterparts due to the limited material selection available. Seeking novel materials with substantially higher energy density seems a promising avenue. The use of pseudo-capacitive materials and heteroatom-doped carbon materials in supercapacitors is an effective way to boost energy density, while alloy and conversion materials, such as silicon with much higher theoretical capacities (4200 mAh g^{-1}) compared with common insertion materials (372 mAh g^{-1} in graphite), are probably the solution for Li-ion batteries.⁴²⁻⁴⁴

Battery systems other than Li-ion batteries are being investigated extensively, specifically Li-S and Li-O₂ batteries, for promising next-generation high energy density EES devices.⁴⁵ Nevertheless, batteries based on emerging systems are at still the embryonic stage; there is a long way to verify their practical role in customizable EES devices. Safety issues residing in current EES systems should not be overlooked when designing next-generation EES devices. It is still unclear about the toxicity and maximum dosage of the electrode materials, as well as the effects of newly-developed nanomaterials on human body.^{10, 46} At the current stage, a practical solution is to employ biocompatible and mechanically robust packaging to protect human body from corrosive and toxic materials. To fully integrate customizable EES devices with human body, natural materials and biofriendly alternatives are required to replace the toxic and corrosive device components.^{10, 47-51} This replacement will make devices more reliable and biocompatible for further ingestible and implantable application. In addition, current corrosive or flammable electrolytes can result in health hazards due to potential electrolyte leakage during the device lifetime. Developing solid-state electrolytes is deemed as most critical to address the safety issue.⁵² Unfortunately, their ionic conductivity and adhesion with electrode materials are still lacking, losing out to conventional electrolytes for practical applications.

Solely improving electrochemical properties is not enough; there are still many challenges in the current electrode and device structures, calling for more deformation-compatible structure designs. For wave-like and fiber-shaped structures, they are only compatible with active materials that are robust enough to form the desired structure. The limited materials selection, mainly CNT and graphene-based materials, restricts the use of active materials with higher capacity and hinders further increase in the performance of customizable EES devices. For the bridge-island structure, the balance between the high plane coverage of active materials and high stretchability of

electrodes is essential yet tricky. Moreover, the mismatch in terms of resiliency between active materials and elastic substrates often results in stress concentration at the interface, leading to performance degradation and long-term instability during the stretch-and-release process.⁵³ Besides, newly developed flexible and stretchable structures are mostly focused on 2D surfaces with limited areal energy density, while the limited surface area of human body requires the development of customizable 3D EES devices with deformability and enhanced areal capacity.³¹ Therefore, there is still a need to devise other ingenious electrode and device structures that are compatible with current materials for fabricating high-performance and highly deformable EES devices. Furthermore, the lack of understanding the interactions in the electrolyte/electrode interface under deformation conditions results in uncontrolled performance in these devices. Studies through in-situ characterization and advanced simulation techniques will provide deep insights into the electrode interface behaviors in customizable EES devices, and will benefit the structure optimization and new structure design.⁵³⁻⁵⁵

Further extension of efficient fabrication methods and integrated multifunctional systems

As expected, the customizability of EES devices would require more complex fabrication processes than their conventional EES counterparts, which may potentially hinder the translation to industrial mass production. A slurry-coating process is usually used to fabricate planar electrodes in conventional supercapacitors and batteries. However, this process is not suitable for fabricating electrodes with different constituents and specially designed structures, which is usually the case in customizable EES devices.^{24, 31, 56-60} Therefore, controllable, scalable and high-throughput manufacture of structured electrodes and devices will remain a research focus for customizable EES devices. Various printing technologies (screen, spray, jet, gravure and 3D printing), laser-writing and holographic patterning (photolithography) growing rapidly in recent

years might be viable for the fabrication of customizable EES devices.⁶¹⁻⁶³ Recently, 3D printers have successfully printed customizable Li-ion batteries with thick electrodes through extruding and then solidifying anode and cathode inks, coupled with the UV curable separator and packaging inks to form arbitrary geometries.⁶⁴⁻⁶⁵ It is feasible to apply these technologies to achieve the customizable structural electrodes and devices with precise, highly-uniform structures.

The ultimate beneficiary of customizable EES devices would be wearable and bio-integrated electronics. The ability to integrate other electronics components or incorporate multifunctionalities into one device is an inevitable route for future customizable EES devices. Although abundant customizable EES device prototypes have been demonstrated, only a few functions have been integrated with the devices. Wireless power transmission presents a key feature to be integrated into customizable EES devices for wireless-charging electronics. Additionally, for self-powered electronics, customizable EES devices should further couple with energy conversion devices (e.g., solar cell, microbial fuel cell, piezoelectric nanogenerators) through harvesting energy from body heat, movement, and ambient light.⁶⁶⁻⁷³ Moreover, through functionalized electrode or electrolyte with advanced materials, functions such as optical transparency, self-healing, electrochromism can also be incorporated.⁷⁴⁻⁷⁵ Other functions including shape-memory, thermal responsiveness, and photodetection are believed to be essential in advanced applications of artificial intelligence and biomimetic technology, which will be explored for smarter and customizable four-dimensional (4D) electronics.⁷⁶

Artificial intelligence enabled programmable combinations of customizable properties

After the first decade of customizable EES devices, we are now entering a period where the combination of customizable properties is important. These customizable properties include but are not limited to energy storage capacity, mechanical flexibility, shape, and functionality,

according to practical needs (Figure 3). However, the current research on customizable EES devices mainly focuses on improving each individual property, which unfortunately compromises the combination of these collective customizable properties. Recent years have witnessed an explosive development of artificial intelligence and their broad-spectrum applications in various scientific areas. Artificial intelligence is becoming a powerful tool in predicting and analysing complicated relationships, which may aid materials scientists to precisely link the relationship between the hierarchical structures (i.e., material, electrode, device and system) and customizable properties (i.e., energy storage, mechanical flexibility, shape, and functionality) of customizable EES device. Making full use of powerful computational capabilities, further studies could explore the following directions: (1) building multiparametric combinational database for computational simulation; (2) recruiting artificial intelligence in the discovery of new customizable EES devices; (3) merging computational simulations with experiments for database verification and analysis.

A multiparametric combinational database would provide us with higher versatility for the customizable combination of properties. Given the complex and interdependent physicochemical properties in the combinational process, an extensive interpretation of the key parameters for the customizable combination is necessary to optimize material-device interfaces and to deliver customizable combinational devices. The intrinsic energy storage capacity of a material is determined by its intrinsic thermodynamic properties of the material, while the real energy storage performance of an EES device is usually biased by some physical parameters (such as size, shape, and stacking structure). Based on these parameters of the electrode materials, the energy storage performance of an EES device can be affected by different pathways, such as the electrode conductivity, ion-transfer pathways, and electrolyte penetration into the electrode. Furthermore, other physicochemical properties (e.g., mechanical, biocompatible, stimuli-responsive properties)

of the materials can also influence the stability of the electrode structure, the feasibility of device fabrication and the expression of the functionality in the device systems. All these need to be factored as weighted parameters when modelling the performance of customizable EES devices. Building a multiparametric combinational database for the computer simulation will contribute to the systematic understanding of the combinational process.

Traditionally, experimental data are first collected, followed by prediction via extrapolation. However, this method is incompatible with a large amount of data for multiparametric prediction. Advances in artificial intelligence or machine learning enable the computers through learning empirical data to discover hidden relationships between inputs and outputs of a system without involving the explicit mechanisms. Considering the complex variables in customizable combination, machine learning can be a viable technique to increase the efficiency and accuracy of the design of a customizable EES device. Recently, artificial neural networks have been used in the prediction of electrochemical performance, including capacitance prediction for carbon-based supercapacitors⁷⁷, the design of Li-ion batteries⁷⁸ and the remaining battery life prediction⁷⁹. Deep learning can also be extended to predict and design the electrochemical and mechanical performance of the material, composite materials, combinational material and structure at multiscale structures of customizable EES devices.

It is noteworthy that through deep learning, the algorithms from machine learning models can permute the combinational properties to find new types of customizable EES devices and predict their physicochemical properties. The incorporation of multiparametric combinational database and artificial intelligence can profoundly accelerate the design and identification of optimal combination process, and effectively narrow down the permutations of the combinational properties for subsequent experimental verification and analysis. Merging computer simulation

and experiments into holistic design tool will reciprocally benefit the translation of conceptual combinational properties of customizable EES devices into controllable and programmable products.

Outlook

The first-decade development of customizable EES devices has seen the fusion of customizable properties. Advances in the materials, electrode structures, device fabrications and system integrations each created the technological building blocks that provide the present EES devices with capacity, flexibility, shape, and functionality. The future of customizable EES devices should create the all-in-one platform, allowing manufacturers and users to program them to perform different functions for different needs in real life. Through novel materials engineering, innovative structure design, and robust computer simulation and artificial intelligence, we might expect the development of customizable EES devices to create a user-friendly and personalized interaction for future wearable and bio-integrated electronics.

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Notes

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Customizable electrochemical energy storage devices

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Customizable Electrochemical Energy Storage Devices

TOC Figure

Quotes

1. The customizable EES devices are stretching the concept beyond capacity and rate capability, to an extent where they are adaptable with wearable and bio-integrated electronics.
2. We are now at a critical point where the design of EES device needs to be intertwined with real-world applications for creating a user-customizable device.
3. Artificial intelligence is becoming a powerful tool in predicting and analysing complicated relationships, which may aid materials scientists to precisely link the relationship between the hierarchical structures (i.e., material, electrode, device and system) and customizable properties (i.e., energy storage, mechanical flexibility, shape, and functionality) of customizable EES device.