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Biomaterials and bioelectronics for self-powered neurostimulation

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ABSTRACT

Self-powered neurostimulation via biomaterials and bioelectronics innovation has emerged as a compelling approach to explore, repair, and modulate neural systems. This review examines the application of self-powered bioelectronics for electrical stimulation of both the central and peripheral nervous systems, as well as isolated neurons. Contemporary research has adeptly harnessed biomechanical and biochemical energy from the human body, through various mechanisms such as triboelectricity, piezoelectricity, magnetoelasticity, and biofuel cells, to power these advanced bioelectronics. Notably, these self-powered bioelectronics hold substantial potential for delivering neural stimulations that are customized for the treatment of neurological diseases, facilitation of neural regeneration, and the development of neuroprosthetics. Looking ahead, we expect that the ongoing advancements in biomaterials and bioelectronics will drive the field of self-powered neurostimulation toward the realization of more advanced, closed-loop therapeutic solutions, paving the way for personalized and adaptable neurostimulators in the coming decades.

1. Introduction

Neurological dysfunctions rank among the leading causes of mortality and diminished quality of life [1–3]. Conditions such as Alzheimer's, epilepsy, amyotrophic lateral sclerosis (ALS), Parkinson's, multiple sclerosis, depressive disorders, sensory system dysfunctions, and various neurodevelopmental disorders exert significant health and economic burdens globally [4–6]. Owing to the electrically excitable nature of neurons, electrical stimulations (ES) have gained traction as therapeutic interventions for these neural dysfunctions [7–9]. Applications of neurostimulation include modulating nerve systems, restoring sensory pathways, and fostering neural regeneration. Particularly, they become indispensable when molecular therapies are hindered by obstacles like the blood-brain barrier [10]. Over the past four decades, Deep Brain Stimulation (DBS) technologies have evolved significantly, offering treatments for disorders including but not limited to Parkinson's, epilepsy, dystonia, essential tremor, obsessive compulsive disorder, and Alzheimer's [11–14]. However, a major limitation of these systems is their reliance on batteries, which, when depleted, necessitate risky surgical replacements [15,16]. The potential risks associated with surgical replacement of batteries make it imperative to explore alternative power solutions. Hence, there is a two-fold necessity: catering to the growing global demand for restoring and repairing dysfunctional nervous systems and resolving technical challenges within existing neurostimulation therapies through innovative approaches for powering neurostimulators.

Neuroengineering, a dynamic and interdisciplinary field bridging neuroscience and engineering, seeks to understand, interface with, and modulate the complex functions of the nervous system (Fig. 1A). Leveraging advanced tools and methodologies from electrical engineering, materials science, and computational modeling, neuroengineers strive to decode neural circuits, develop neuroprosthetics, and forge innovative treatments for neurological disorders [17–20]. This intersection of biology and technology not only unravels insights into the intricate workings of the brain but also heralds transformative solutions that can restore or enhance neural functions, ultimately ameliorating the lives of individuals with neurological conditions [21–23]. Engineering solutions have diversified the potential applications in neurostimulation, such as deep brain stimulation, vagus nerve stimulation, retinal stimulation, transcranial magnetic stimulation, sacral nerve stimulation, sciatic stimulation, and spinal cord stimulation (Fig. 1A). The landscape of neuroengineering has been greatly enriched by the emergence of self-powered bioelectronics and biomaterials. Recent advancements in this area offer a viable alternative for energy harvesting, which can power neurostimulators and other neuroengineering applications [24–26]. Parallel to the rapid development of

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the Internet of Things (IoT), these devices have diversified their potential applications, emphasizing aspects like flexibility, wearability, and safety [27]. These bioelectronics have the capability to harness energy from different sources, including body biomechanical motions and organ activities, for miscellaneous neurostimulation applications (Fig. 1A) [28–32]. Cutting-edge platform technologies like triboelectric nanogenerators (TENGs) [33–35], piezoelectric nanogenerators (PENGs) [36–38], magnetoelastic generators (MEGs) [28,39,40], and biofuel cells [41–49] possess the ability to convert either biomechanical or biochemical energy to electricity to interface with the nervous systems (Fig. 1B).

In this review, we will present a comprehensive overview of the latest developments in self-powered neurostimulation via biomaterials and bioelectronics innovation. We will expound on the working mechanisms of these neurostimulation devices, highlighting their applications in stimulating various populations of neurons in both the central and peripheral nervous systems. Additionally, we will emphasize the profound impact of these technologies in shaping personalized, wearable treatments. We will also explore the untapped potential of these bioelectronics in the context of neural regeneration and neuroprosthetics. Finally, we will discuss their prospective growth of the neuromodulation market and subsequent impact brought to its research. As well as drawbacks in materials of concurrent self-powered devices and their integration into future intelligent closed-loop therapeutic systems for the nervous system care.

2. Biomaterials used in self-powered bioelectronics for neuroengineering

The evolution of neuroengineering has been significantly influenced by the development and integration of advanced biomaterials into self-powered bioelectronics [50,51]. These materials not only enhance the device's efficiency and functionality but also guarantee biocompatibility, a key consideration for any implantable or wearable devices [52,53]. This section examines the variety of biomaterials that are commonly used in self-powered bioelectronics with a focus on their relevance in neurostimulation. The primary self-powered techniques that have gained traction in the realm of neuroengineering include TENGs, PENGs, MEGs, and biofuel cells. Each method harnesses energy from diverse sources, whether biomechanical motions, magnetic fields, or biological reactions, and transforms them into electrical power for a range of neuroengineering applications [54–59]. Applications of these self-powered techniques extend across various sensory mechanisms and therapeutic interventions, from the perception of touch and pressure to neural repair and regeneration, which focuses on restoring damaged neural pathways and functions, as shown in Table 1. Further details about each of these techniques will be discussed in the sections that follow.

2.1. Triboelectric nanogenerators

Implantable bioelectronics have undergone notable advancements, particularly in harnessing renewable energy sources from the human body [60,63,66,67,93–101]. Triboelectric nanogenerators (TENGs), which translate mechanical movements into electric energy by using the triboelectrification between two dissimilar materials, are at the forefront of this development [102–105]. The choice of materials is vital for the effectiveness of TENGs. This section delves into the materials employed in TENGs, emphasizing their critical role in biocompatibility, flexibility, and efficiency.

2.1.1. Materials for triboelectric layers

Materials for triboelectric layers fabrication can be divided into organic and inorganic types, as follows.

Organic Materials: ***Polydimethylsiloxane (PDMS)*** is a compelling choice for TENG substrates owing to its biocompatibility, mechanical flexibility, and straightforward fabrication process. PDMS not only is biocompatible but also minimizes the risk of adverse reactions when interfaced with human tissues. Its attributes make it an excellent choice for a wide array of biomedical applications. ***Teflon*** (i.e., polytetrafluoroethylene (PTFE) and fluorinated ethylene propylene (FEP)) is renowned for its unique negative triboelectric properties, which are instrumental in electrical charge generation in TENGs. When coupled with a compatible material, Teflon facilitates effective energy conversion, a critical component for implantable devices. ***Biodegradable polymers*** (i.e., polylactic acid (PLA) and polyglycolic acid (PGA)) are well-suited for short-term implants, and their biodegradable nature allows them to be adapted for TENGs based soft bioelectronics and naturally decompose within the body. ***Hydrogels*** are biocompatible, water-absorbent polymers with mechanical properties that closely mimic biological tissue, making them ideal candidates for soft bioelectronics that require a tissue-mimetic interface.

Inorganic Materials: ***Metal foils*** (i.e., aluminum, gold, copper) act as electrodes, enhancing charge collection and transport in TENGs. They

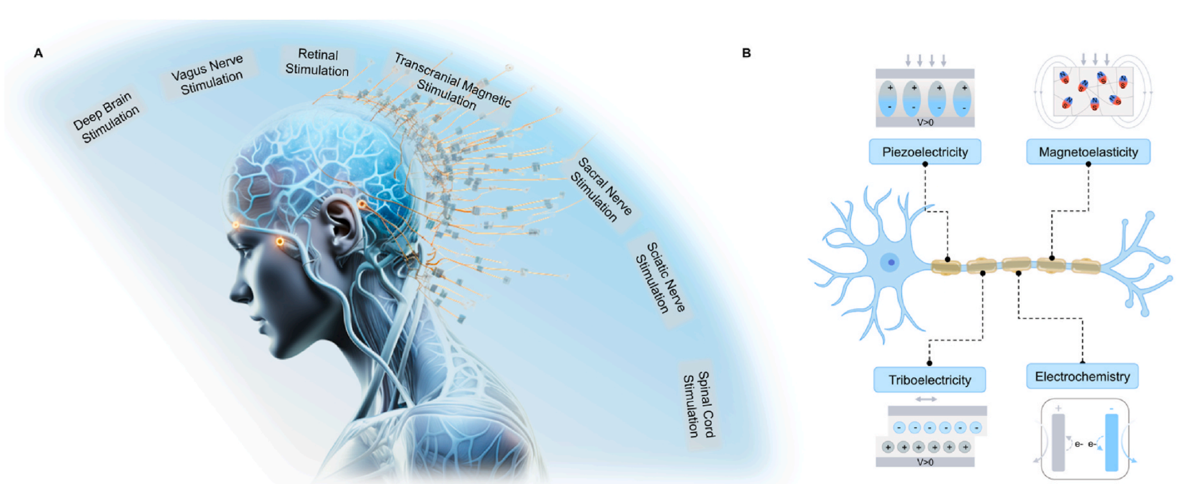

Fig. 1. Overview of the interplay between biomaterials and bioelectronics for neuroengineering applications. (A) Delineation of targeted application areas for self-powered neuroengineering devices, deep brain stimulation, retinal stimulation, vagus nerve stimulation, sacral nerve stimulation, transcranial magnetic stimulation, spinal cord stimulation, and sciatic nerve stimulation. (**B**) Categorization of the various self-powered techniques examined in this paper, which include piezoelectric, magnetoelastic, triboelectric, and electrochemical modalities.

Table 1
Summary of materials used for neuroengineering applications.

Method	Application	Material	Ref.
TENG	Tactile Sensing	PDMS, Au, Kapton, VHB	[60]
		PDMS, Cu, Ag, ZnS	[61]
	Visual Sensing	Ppy, Cu, PDMS	[62]
	Olfactory Sensing	Ppy, Cu, PDMS	[63]
		PET, PTFE, PANI	[64]
	Auditory Sensing	Acrylic, Kapton, Au, FEP	[65]
	Brain Stimulation	PDMS, Teflon, ITO, PET	[66]
	Vagus Nerve Stimulation	Ecoflex, PDMS, Cu, PTFE	[67]
		PI, Si-PU, PTFE, Ag, PVDF	[68]
	Neurogenic Unreactive Bladder Stimulation	NiTi, PVC	[69]
		PET, PTFE, Al	[70]
	Sciatic Nerve Stimulation	Ecoflex, PDMS, Al, Ag, PA6	[71]
		Cu, PDMS, PET	[72]
		PDMS, ITO, PET	[73]
	Neural Repair and Regeneration	PLGA, MG, PCL, PVA	[74]
		Al, Cu, PDMS, Kapton	[75]
		PMMA, Cu, Kapton, Al	[76]
PENG	Tactile Sensing	Au, PVDF	[77]
	Auditory Sensing	PZT	[78]
		$BaTiO_3@SiO_2$	[79]
	Brain Stimulation	PIMNT, Au, PET	[80]
		PDMS, Sm-PMN-PT, Au	[81]
		PZT, PDMS	[82]
		PZT, Cu	[80]
		PZT, PVDF	[83]
	Vagus Nerve Stimulation	PI, PVDF, Au	[84]
	Neural Repair and Regeneration	ZnO/PCL	[85]
		PVDF	[86]
		PVDF	[87]
		PVDF/PVC	[88]
MEG	Neural Repair and Regeneration	ITO, PDMS, Cu, NdFeB	[28, 89]
Biofuel Cell	Brain Stimulation	CNT, GO_x, BOD	[90]
		AuNPs, PL	[91]
	Neural Repair and Regeneration	GOX, BOD, EBFC	[92]

offer the stability and reliability that are crucial for implantable bioelectronics. ***Magnesium alloys*** are also noteworthy due to their biocompatibility and biodegradability, making them suitable for temporary implantable TENGs. They dissolve naturally in the body, leaving no residue. ***Inorganic nanostructures*** (i.e., zinc oxide, copper oxide, silver) are employed in TENGs to amplify triboelectric performance. These nanostructures, given their expansive surface area, can be integrated into flexible substrates for specialized implantable applications.

2.1.2. Electrode materials used in TENGs

Metals: ***Gold (Au)*** electrodes are commonly used in a variety of biomedical applications owing to their excellent conductivity, chemical stability, and biocompatibility. They often serve as the primary electrode in TENGs [96–98]. ***Copper (Cu)*** electrodes are valued for their superb electrical conductivity, facilitating efficient current flow. However, their use comes with caveats. While copper is generally considered safe for internal use, it is prone to corrosion over time. This corrosion may lead to the release of copper ions, which could pose health risks. To mitigate this, coatings or encapsulation methods are often employed to ensure safety [98,99].

Other Materials: ***Graphene***: Comprising a single layer of carbon atoms in a hexagonal arrangement, graphene boasts remarkable electrical conductivity and mechanical resilience. Electrodes based on graphene have been investigated for use in flexible TENGs [100]. ***Indium tin oxide (ITO)*** serves as an efficient conductor, ensuring optimal charge transport in TENGs, which is critical for device performance [101]. ***Conductive polymers*** (i.e., polyaniline (PANI), polypyrene (PPy), and poly (3,4-ethylenedioxythiophene) (PEDOT)) are also utilized as electrode materials in flexible and implantable electronics. These polymers can be molded into forms that are both flexible and biocompatible, extending their potential uses [101].

2.2. Piezoelectric nanogenerators

Piezoelectric nanogenerators (PENGs) represent a compelling technology for implantable bioelectronics. These devices leverage mechanical movements to generate electrical energy, and their small size, high efficiency and robustness enable the development of cutting-edge, durable biomedical implants [102]. Such progress is not merely incremental; it opens new horizons for patient care and healthcare technology as a whole [103].

2.2.1. Piezoelectric materials

Piezoceramics such as lead zirconate titanate (PZT) is a prominent piezoceramic material used in implantable bioelectronics, attributed to its excellent piezoelectric properties. It is efficient at converting mechanical strain into electrical energy. However, its lead content poses serious questions regarding biocompatibility and environmental impact [104]. ***Leadless piezoceramics*** alternatives like potassium-sodium niobate (KNN) ceramics, barium titanate (BaTiO3), and sodium bismuth titanate (NBT) are emerging as substantial candidates in implantable bioelectronics. These lead-free materials mitigate the environmental and health concerns of traditional piezoceramics [105]. ***Piezoelectric polymers*** like polyvinylidene fluoride (PVDF) and its derivatives are the preferred piezoelectric polymers in PENGs due to their adaptability during the fabrication process [106]. ***Biodegradable piezoelectric materials*** including poly (L-lactic acid) (PLLA), poly (glycolic acid) (PGA), its copolymers, and polyhydroxyalkanoates (PHA). These materials not only produce electricity from mechanical stress but also naturally degrade over time, rendering surgical removal unnecessary and providing an eco-friendly alternative for implantable devices [107].

2.2.2. Associated components and materials

Biocompatible substrates, including materials like silicon and flexible polymers (i.e., polyimide), act as foundational layers in PENGs. Silicon offers structural stability, whereas flexible polymers conform to the body's shape. This adaptability enables seamless integration of PENGs into a variety of implantable devices [108]. ***Electrode materials***, essential to the function of PENGs, are tasked with collecting and channeling electrical charge. Noble metals such as gold and platinum are frequently chosen as electrode materials due to their high conductivity and resistance to corrosion.

Biocompatible encapsulation is crucial for ensuring the durability and safety of PENGs when implanted within the body. Commonly employed materials for this encapsulation include PDMS, polyurethane, and medical-grade silicone [109]. These materials are specifically selected due to their biocompatible properties and effectiveness in providing a robust protective layer around PENGs.

2.3. Magnetoelastic generators

Magnetoelastic generators (MEGs) are an emerging and fundamentally new platform technology that was firstly invented in 2021 [28] and designed to convert mechanical pressure into electrical signals by utilizing magnetoelastic effect in soft materials systems [89]. These materials exhibit a change in their magnetic properties in response to mechanical stresses or strains. When subjected to vibrations or deformations, the materials experience shifts in their magnetic attributes, thereby generating an electrical voltage in the soft conductive coils owing the electromagnetic induction. A typical magnetoelastic generator consists of two primary components: a magnetomechanical coupling (MC) layer and a magnetic induction (MI) layer.

2.3.1. Magnetic coupling (MC) layer

The MC layer in a magnetoelastic generator is composed of a polymer substrate mixed with magnetic nanoparticles. When uniformly mixed, these components form a soft magnetoelastic composite system.

PDMS according to a prototype design by *Zhou* et al., acted as the polymer substrate, while ***NdFeB*** was utilized as the nanomagnetic particle [28]. The MI layer in a magnetoelastic generator is fabricated using a soft liquid metal coil system. It aims to convert biomechanical motions into magnetic flux density variations, which is then translated into electricity by the MI layer. Although various types of polymer substrates and magnetic nanoparticles can be integrated into the MC layer due to its straightforward structural design, biocompatibility remains a crucial factor, especially for implantable applications, which necessitating materials like PDMS. Advances in coating and encapsulation technologies, such as silica coating on the magnetic nanoparticles, are widening the array of materials that can be used in MEG designs with improved biocompatibility.

2.3.2. Magnetic induction (MI) layer

The MI layer in a magnetoelastic generator is responsible for capturing magnetic flux density fluctuations and converting these magnetic changes into electrical energy, relying on electromagnetic induction. The design incorporates soft coils specifically engineered to harvest energy from the generated magnetic field. To meet the stringent requirements of flexibility and stretchability for wearable and implantable bioelectronics, coils are often uniquely shaped and are made from soft ***copper fibers*** or from more flexible substances like ***liquid metal***. Liquid metal, when used as an MI layer material, has multiple advantages: it is inherently flexible and stretchable, thus works as an ideal choice for wearable applications requiring adaptability to body movements. In addition, gallium-based liquid metals offer excellent electrical conductivity and biocompatibility, making them well-suited for future magnetoelastic generator designs [110].

2.4. Biofuel cells (BFCs)

Biofuel cells (BFCs) have been gaining attention as a pivotal technology for constructing bioelectronic devices, offering a sustainable and renewable energy source for a variety of medical applications. Unlike conventional traditional batteries that require frequent replacement or recharging, BFCs generate electricity through biochemical reactions, making them ideal candidates for self-powered neurostimulation. This section will explore the two primary components of BFCs: the anode and cathode [111].

2.4.1. Anode materials

Carbon-based materials (i.e., Carbon nanotubes, graphene) are popular choices due to their expansive surface area, superior electrical conductivity, and biocompatibility. They provide a robust platform for enzyme adhesion, which in turn facilitates electron transfer and enhances the anode's catalytic activity.

Nanocomposites like carbon nanotube-polymer composites offer both mechanical resilience and electrical conductivity, features that are beneficial for enhancing enzyme adhesion and facilitating efficient electron transfer. ***Enzyme-modified materials*** are often tailored with enzymes such as glucose oxidase or dehydrogenase to maximize anode performance. These enzymes are anchored onto conductive substrates, like carbon nanotubes, using various techniques ranging from cross-linking and adsorption to covalent bonding.

2.4.2. Cathode materials

Platinum (Pt) and its alloys are well-known for their catalytic efficiency in oxygen reduction reactions and are commonly used in cathodes. However, the high cost and potential long-term toxicity of Pt make it a material that requires thorough evaluation, particularly for implantable devices. ***Carbon-based substrates*** such as carbon cloth or carbon paper are also employed in cathodes. They act as structural backbones for catalysts and aid in the oxygen reduction reaction [112]. ***Enzymes and Organisms*** are innovative approaches including the use of enzymes (i.e., laccase) or microorganisms (i.e., specific bacteria), which can directly facilitate oxygen reduction. This design directly facilitates Oxygen reduction while eliminating the need for expensive metal catalysts and offering a biocompatible alternative [113].

3. Platform technologies for self-powered neurostimulation

3.1. Triboelectric nanogenerators (TENGs)

3.1.1. Working principles

The triboelectric effect describes the phenomenon where dissimilar materials acquire an electric charge after making frictional contact [114–117]. This effect is a ubiquitous electrostatic occurrence, originating from the contact and ensuing separation of different materials. The relative movement between two charge layers results in an electric potential difference in between. The triboelectric series ranks materials based on their likelihood to either gain or lose electrons during such interactions [118]. Materials positioned higher in the series tend to lose electrons and acquire a positive charge, whereas those lower in the series usually gain electrons, leading to a negative charge. Optimized for both electrical output and biocompatibility, TENGs convert biomechanical movements into electrical energy, making them highly suitable for powering neurostimulation devices. Various methodologies, encompassing physical, chemical, and hybrid techniques, have been employed to enhance these nanogenerators' electrical outputs. The TENGs predominantly employ four working mechanisms: vertical contact-separation, lateral sliding, single-electrode, and freestanding triboelectric-layer modes [119–121]. These operational modes are briefly explained as follows: in vertical contact-separation mode, charges are generated when materials come into perpendicular contact, as illustrated in Fig. 2A [122–128]. Conversely, the lateral-sliding mode generates electricity through horizontal movements between two materials without the need for separation [129–133]. In the single-electrode mode, one electrode is grounded while the other generates electricity through motion [72,73,133–135]. Lastly, the freestanding triboelectric-layer mode involves the movement of a triboelectric layer relative to two electrodes, generating charge via electrostatic induction [136,137].

3.1.2. Applications in self-powered neurostimulations

Since the inaugural report of TENG in 2012, TENGs have found applications in a wide range of wearable biomedical applications, including drug delivery, muscle stimulation, and health monitoring [119], such as drug delivery [139–141], muscle stimulation [61,62,142–144], health and disease monitoring [64,65,69,145,146]. In recent years, several studies have demonstrated TENG's efficacy in interfacing with the nervous system. These applications span from sensory nervous system restoration to therapeutic stimulation and neural repair. Dysfunctions in the sensory nervous system manifest in various forms, such as visual impairments, hearing loss, ageusia, anosmia, and tactile dysfunctions. TENG offers self-powered solutions to these challenges through electrical stimulation. For example, *Fu* et al. designed a TENG-based multi-perception receptor for sensory substitution that covered multiple senses such as touch, hearing, smell, taste, and sight. This innovation was proven to affect mouse behavior when integrated into the S1BF cortex (Fig. 2B and C) [98]. In terms of visual restoration, *Dai* et al. crafted an artificial retina using a Ppy/PDMS triboelectric-photo-detecting matrix that was energized by natural human movements like blinking (Fig. 2D) [70]. This e-skin could detect both single-point and multi-point UV illumination, driven by human activity. Furthermore, the system could transmit perceptual signals and

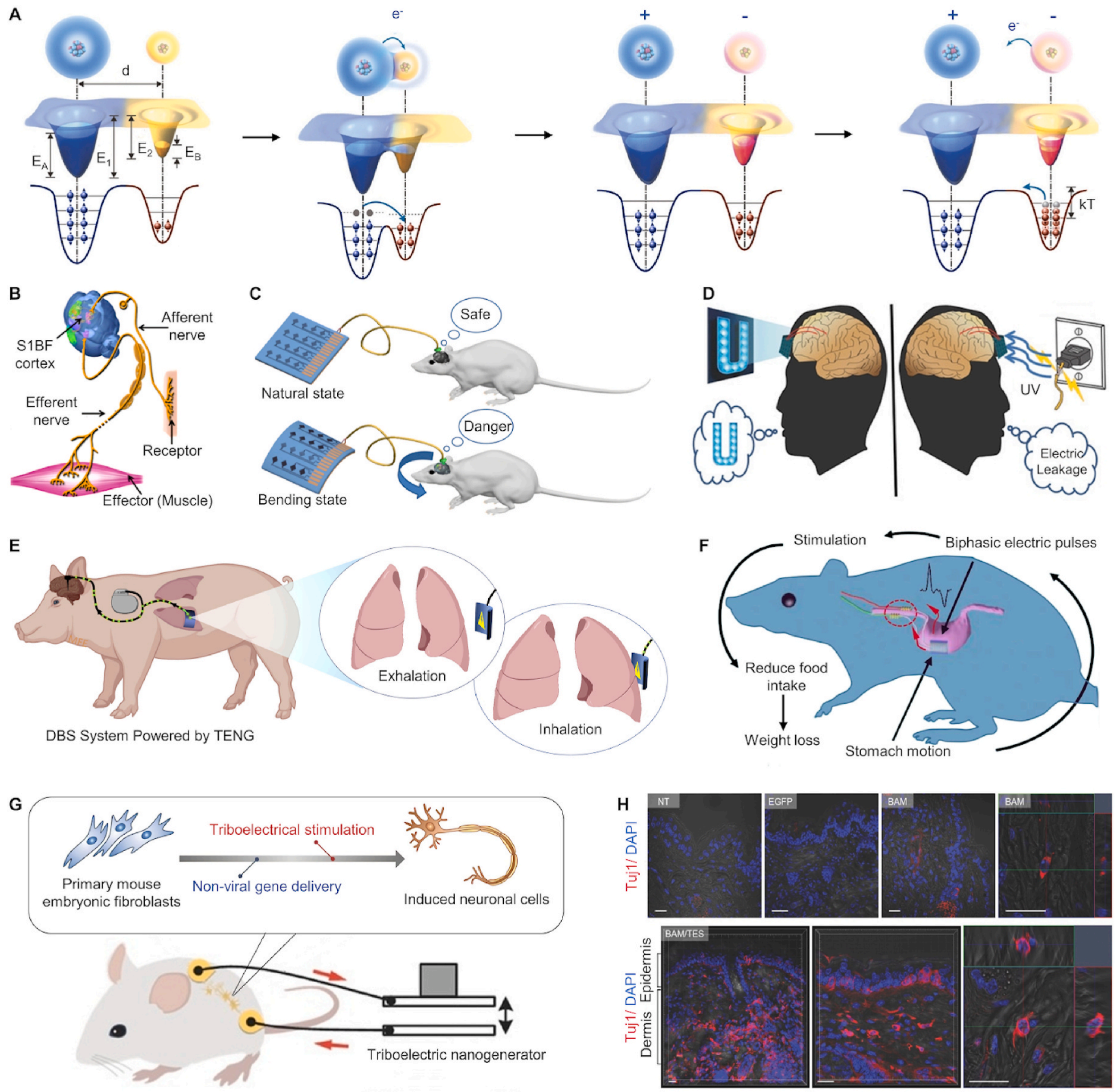

Fig. 2. Working principles of TENGs and their applications in neurostimulation. The primary objectives include: 1) the restoration of sensory functions, 2) the administration of neuromodulation therapeutics, and 3) the repair and regeneration of the nervous system. (A) Illustrates the triboelectric charge transfer process utilized in the contact-separation working mode [138]. (A) Reproduced with permission. Copyright 2018, Wiley-VCH. (B) Details the neural pathways in the S1BF region relevant to sensory perception [93]. (B) Reproduced with permission. Copyright 2018, Elsevier. (C) Describes the methodology adopted for modulating mouse behavior and inducing threats through TENG-powered neurostimulations [93]. (C) Reproduced with permission. Copyright 2018, Elsevier. (D) Depicts a schematic of a Ppy/PDMS-based TENG that is designed for detecting illumination and transducing signals to the brain [128]. (E) Features a schematic that outlines the use of TENG in powering a deep brain stimulator in swine, which is activated by periodic exhalation and inhalation of the lungs [66]. (F) Showcases the architecture of a TENG-empowered vagus nerve stimulator, operated by movements of the stomach, with the aim of controlling body weight [67]. (G) Portrays how TENG-assisted electrical stimulation facilitates the conversion of fibroblasts to neurons while working alongside the non-viral delivery of genes encoding cell lineage-specific transcription factors (TF) [139]. (G) Reproduced with permission. Copyright 2016, Wiley-VCH. (H) Exhibits the immunostaining outcomes of mouse skin tissues in NT, EGFP, BAM (top), and TES/BAM condition groups (bottom) as detected by anti-Tuj1 antibody [139]. (H) Reproduced with permission. Copyright 2016, Wiley-VCH.

deliver illumination to the mouse brain across a range of intensities and wavelengths. TENGs have also been used in restoring tactile sensations [98]. *Zhang* et al. reported a self-powered triboelectric-mechanoluminescent electronic skin (STMES) capable of differentiating various mechanical stimuli such as stretching, bending, and pressing through triboelectric-induced electrical signals and mechanoluminescence optical signals [147]. *Shlomy* et al. developed an integrated tactile TENG (TENG-IT) system that could integrate tactile information with the dorsal ganglion root, primarily responsible for sensory transduction, in mice *in vivo* [96]. *Yu* et al. created a TENG-based system for detecting tactile stimuli including displacement, pressure, and touch patterns, and transduced these signals to artificial afferents for spatiotemporal recognition [68]. This system serves as a valuable framework for biomimetic and neuromorphic electronics that process intricate tactile data, capable of integration with neural afferents for delivering sensory information to the brain. TENGs also assist in the transduction of olfactory signals. *Zhong* et al. developed a TENG-powered PDMS/Ppy olfactory substitution system that could differentiate between various gas species *in vitro* and modify mice' behavior through delivering triboelectric currents to the mice' barrel cortex *in vivo* [99]. *Xue* et al. designed a PANI/PTFE/PANI system combining triboelectrification with a gas-sensing unit for discriminating ethanol odor concentrations in the air [71]. Additionally, *Guo* et al. designed triboelectric auditory sensors (TAS) capable of distinguishing soundwaves. The TAS utilized an architecture of Acrylic/Kapton/Au/FEP and demonstrated to be able to discern sound frequencies between 100 Hz and 5000 Hz, showing potential for auditory implant applications [74]. In summary, contemporary TENG designs have successfully transduced electrical signals across various sensory modalities in the brain.

Recent advancements in TENG-based soft bioelectronics have contributed to neuromodulation therapies affecting both the central and peripheral nervous systems. These devices harness energy from the rhythmic movements of internal muscles and organs, such as the stomach, bladder, and lungs, to stimulate adjacent nervous tissues. Commercially available neurostimulators like deep brain stimulators and spinal cord stimulators have been improved through these advancements. *Elsanadidy* et al. designed a deep brain stimulator system integrated with a high-performance bio-triboelectric nanogenerator (Bio-TENG) and a rapidly changing super-capacitor [100]. Powered by the inhalation and exhalation cycles in swine, the system achieved a peak power of 6.9 μW and successfully stimulated mouse hippocampal cells *ex vivo* (Fig. 2E). *Xu* et al. proposed a nanofibrous-membrane-based TENG as a sustainable treatment alternative for spinal cord injuries (SCIs) [75].

Various TENG designs have been developed for stimulating the peripheral nervous system. For instance, TENG-based neurostimulation has proven effective in treating bladder dysfunctions like underactive bladder by targeting the autonomic pelvic nerve. *Hassani* et al. introduced a bistable microactuator integrated with a TENG sensor [76]. Comprising layers of PDMS, copper, polyethylene terephthalate (PET), and polyvinyl chloride (PVC), this sensor transformed bladder volume changes during filling into triboelectric signals, achieving a voiding efficiency of up to 78 % in mice. *Lee* et al. refined this with a PTE/PTFE/Al TENG-based system coupled with a flexible neural interface clip [148]. The system generated charge-balanced biphasic waveforms from TENG movements and investigated optimal TENG stimulation frequencies for bladder contraction and micturition.

TENGs have also been used in vagus nerve stimulation, affecting a range of physiological functions. *Yao* et al. developed a TEL/PTFE/BEL TENG-based vagus nerve stimulator (VNS) powered by stomach motion [149]. The device sent biphasic pulses to the mice's vagus nerve *in vivo*, resulting in a 38 % reduction in body weight compared to control groups. *Sun* et al. introduced a hybrid nanogenerator (H-NG) for low-level vagus nerve stimulation (LL-VNS) in atrial fibrillation (AF) treatments [150]. This innovative device combines piezoelectric and triboelectric elements and showed a 90 % reduction in AF duration in mice.

Furthermore, TENGs have been investigated for their potential in sciatic nerve stimulation to restore motor functions. *Zhou* et al. designed an implantable self-regulated neural electrical stimulation (ISR-NES) system [151]. Utilizing a PDMS/PA6-based TENG, the system harvested energy from abdominal respiration movements and sent biphasic pulses to a nerve cuff electrode targeting the sciatic nerve in mice. After a period of four weeks, mice equipped with ISR-NES displayed significant improvement in the Sciatic Nerve Functional Index (SNFI). *Lee* et al. introduced a TENG-driven flexible sling electrode system targeting the sciatic nerve, which induced movements in the gastrocnemius medialis (GM) and tibialis anterior (TA) muscles [142]. The TENG component employed a multilayer stacked Cu/PDMS-based design with zigzagged PET layers serving as mechanical support. Although the TENG could harvest energy through hand-tapping with a peak-to-peak voltage output of 160V and short circuit current of 6.7 μA, these outputs were inadequate to meet the current amplitude (∼1 mA) required for existing neurostimulators. Subsequent research focused on maximizing voltage output and stimulation efficacy. *Lee* et al. refined their previous design by introducing a water/air-hybrid TENG (WATENG) [143]. Their updated design employed the triboelectrification between ITO and PDMS, with a water-soaked sponge array situated underneath. They also introduced a force-sensing multi-pixel array and a multi-channel sling electrode for delivering exponential electric pulses more precisely, enabling more complex limb movements such as plantar flexion (PF) and ankle dorsiflexion (DF). *Wang* et al. developed a diode-amplified TENG (D-TENG) to increase the triboelectric current for sciatic neurostimulation [144]. The device was constructed in a zigzag stack-layer fashion with Al/PTFE as triboelectric surface pairs. Specifically, the diode architecture achieved a much higher current output during the release stage of the TENG. Meanwhile, J. *Wang* et al. further optimized TENG stimulation protocols to enhance system efficiency and stability [62]. They improved the stimulation efficiency by redesigning the electrode configuration relative to the force-sensing array. They also utilized longer pulse widths and lower current amplitudes to enhance stimulation stability, ensuring synchronization did not occur with motor neuron recruitment.

In recent years, growing evidence has pointed to the considerable potential of TENG's application in neural regeneration, as well as in addressing neurodevelopmental dysfunctions and neural injuries. Empirical research has consistently confirmed the effectiveness of TENG-based stimulation devices in the restoration of neural tissue, especially in the peripheral nervous system. For example, *Zheng* et al. constructed a biodegradable TENG (BD-TENG) from patterned biodegradable polymers (BDP) like PLGA, PHV/B, PHV, and PCL to navigate neuron growth [152]. This device was then encapsulated in Mg and PLGA, and it produced an open-circuit voltage of 40V and a short-circuit current of 1 μA. After applying a 10 V/mm electric field (EF) at a frequency of 1 Hz to cultured primary neurons for 30 min daily over a span of 5 days, the neurons oriented themselves in alignment with the EF direction. *Jin* et al. discussed TENG's role in converting fibroblasts to functional neurons by employing nonviral delivery methods for cell lineage-specific transcription factors (TFs) (Fig. 2G) [77]. The TENG device in this study utilized aluminum (Al) and pillar patterned PDMS as the triboelectric layers, enclosed by Kapton and copper (Cu). After introducing the TFs Brn2, Ascl1, and Myt1l—collectively known as BAM TFs—into primary mouse embryonic neurons (PMEN) via electroporation, they administered a triboelectric stimulation (TES) with an open-circuit voltage of 30 V and a short-circuit current of 270 nA. Notably, the administration of TES followed the electroporation process. On days 9 and 12–14, the peak number of induced neurons (iN) was noted in the BAM/TES group, as evidenced by the expression of the neuronal marker class III beta-tubulin (Tuj1). *In vivo* experiments involving the transfection of mouse dermal fibroblasts with C32–122 poly (β-amino ester) (PBAE) nanoparticles, which were complexed with BAM TF-expressing plasmids, demonstrated that the BAM/TES combination yielded the highest number of Tuj1-positive cells. This suggests an enhanced efficiency in direct cell conversion (Fig. 2H).

Moreover, *Guo* et al. developed a system employing TENG-driven electrical stimulation to induce the neural differentiation of mesenchymal stem cells (MSCs) [153]. The system utilized PMMA/Kapton layers encapsulated by Cu and Al electrodes and generated outputs of 250V and 30 μA. Neural scaffolds hosting MSCs were fabricated from either 15 % poly (3,4-ethylenedioxythiophene) (PEDOT)–reduced graphene oxide (rGO) hybrid microfiber or rGO microfiber. The MSCs either received TENG-driven electrical stimulation or did not. Results from the *in vitro* experiments revealed a higher differentiation rate into neural stem cells for MSCs on a 15 % PEDOT-rGO scaffold that received TENG-driven electrical stimulation, as confirmed by high expression levels of Tuj1 and glial fibrillary acidic protein (GFAP). Overall, the synergistic application of TENG with other biological interventions has demonstrated its efficacy in promoting neural cell growth, underscoring its transformative potential in future neural regeneration endeavors.

3.2. Piezoelectric nanogenerators (PENGs)

3.2.1. Working principles

Piezoelectric nanogenerators (PENGs) are specialized devices that leverage piezoelectricity, a phenomenon limited to specific materials known as piezoelectric materials, to harvest mechanical energy. Piezoelectricity refers to the capacity of piezoelectric materials to induce inner polarization when subjected to mechanical stress. When external strain is applied to a PENG, its constituent piezoelectric materials, which possess a noncentrosymmetric crystal structure, undergo deformation, thereby disrupting the central symmetry of charge distribution (Fig. 3A). This deformation results in the displacement of positive and negative charges within the material. As different charges accumulate on opposite surfaces, a potential difference across the material arises, allowing electrical output to be captured via electrodes affixed to these surfaces, which can subsequently power an external load. In this way, a continuous electrical output is produced through cyclic deformation of the PENG. The strength of the piezoelectric current generated is contingent on the piezoelectric coefficients and the level of applied stress. The PENGs have been utilized in various biomedical applications including but not limited to neurostimulation, cardiac pacemaking, health monitoring, and tissue engineering [78–80,82,154]. PENGs also have the capability of harvesting mechanical energy from human body motion, such as rotation, twisting, and bending, and the biomechanical activities of the organs, such as the heartbeat, stomach movement, and many others [103].

3.2.2. Applications in self-powered neurostimulations

Like TENGs, PENGs and an array of piezoelectric materials have found extensive applications in neurostimulation for purposes like restoring sensory functions, administering neuromodulatory therapies, and aiding neural regeneration. These devices can effectively emulate sensory signals that are transmitted into the central nervous system, such as tactile or auditory information, due to their distinct physical properties. Chun et al. devised a mechanoreceptor system that couples piezoelectric films and artificial ion channels to mimic the fast-adapting (FA) and slow-adapting (SA) mechanisms found in human skin [81]. These cutaneous receptors in human skin primarily work in two ways: FA and SA [83]. FA receptors, such as Meissner corpuscles (MC) and Pacinian corpuscles (PC), fire rapidly to the stimuli with diminished response for extended presentation of the stimuli. SA receptors, like the Merkel disk (MD) and Ruffini cylinder (RC), respond tonically if the stimuli are present. The sensor system is composed of Au/PVDF piezoelectric film for transducing FA signals, along with PANI as the electrolyte and a porous MB membrane for SA signal transduction, all supported by an Al/C electrode. The system exhibited exceptional sensitivity in both static and dynamic pressure responses (SA, -2.1×10^{-1} V/kPa at 1 Hz and a resting potential of 670 mV; FA, 3.8×10^{-1} and -3.49×10^{-1} V/kPa for two consecutive peaks). The experimental results confirmed the device's ability to sense human blood pressure, distinguish various stimuli (i.e., twisting, rubbing, and bending), differentiate material textures (i.e., glass, Whatman paper, and abrasive paper), and accurately identify Braille characters. *Lee* et al. proposed an

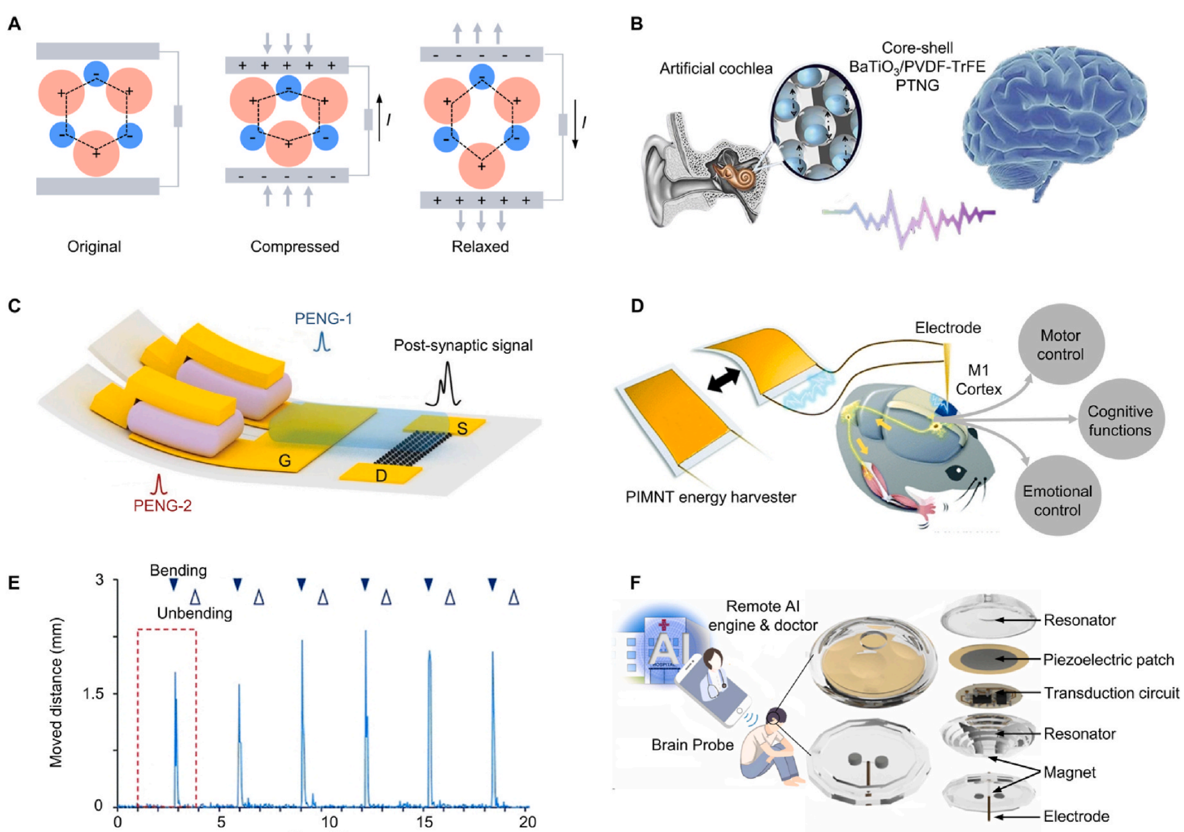

Fig. 3. Working principles of PENGs and their applications in neurostimulation. (A) Illustrates the basic mechanism of a PENG under the influence of external pressure. (B) Features a schematic representation of a core-shell BaTiO3/PVDF-TrFE PTNG capable of transmitting auditory signals directly to the brain [79]. (B) Reproduced with permission. Copyright 2021, ACS. (C) Exhibits a diagram detailing a piezotronic graphene-based artificial neural network designed to mimic the process of synaptic integration [154]. (C) Reproduced with permission. Copyright 2019, Wiley-VCH. (D) Showcases the application of a PIMNT-based PENG in the realm of neural stimulation [148]. (D) Reproduced with permission. Copyright 2015, Royal Society of Chemistry. (E) Quantifies the degree of limb movement achieved through electrical stimulation, which is powered by the flexing and relaxing motions of the PENG [148]. (E) Reproduced with permission. Copyright 2015, Royal Society of Chemistry. (F) Unveils a structural diagram for an innovative brain probe, engineered to enable intelligent, remote neuromodulation therapies [82]. (F) Reproduced with permission. Copyright 2022, Elsevier.

inorganic-based piezoelectric acoustic nanosensor (iPANS) that mimics the functions of human hair cells [155]. Their study included a custom-designed trapezoidal silicon-based membrane (SM) that emulates the human basilar membrane's frequency-selective properties. The iPANS was transferred onto the SM using laser lift-off (LLO) technology. Benchtop evaluations revealed that iPANS converted sound-induced vibrations of the SM at varying frequencies (500 Hz, 600 Hz, and 100 Hz) into distinct piezoelectric voltages (54.8 μV, 46.6 μV, and 59.7 μV), validating its potential for auditory implant applications. Incorporating both triboelectric and piezoelectric mechanisms, *Zheng* et al. crafted a piezo-triboelectric nanogenerator (PTNG) suitable for artificial cochlea application (Fig. 3B) [84]. Their design featured a dual-function core-shell structure: a $BaTiO_3$ core vibrating within a PVDF-TrFE shell. The design allows for the generation of triboelectric charges through contact-separation between the shell and the core, while piezoelectric charges are produced as the core deforms the shell. Using optimized parameters—a composite membrane with a diameter of 30 mm, pore diameter of 276.06 nm, PVDF-TrFE mass ratio of 75 %, and a thickness of 0.1 mm—the core-shell $BaTiO_3$/PVDF-TrFE PTNG achieved an open-circuit voltage of 15.24 V and a short-circuit current density of 9.22 mA m^{-2}. Furthermore, the core-shell $BaTiO_3$/PVDF-TrFE PTNG displayed a robust capability to discern acoustic frequencies ranging from 0 to 1000 Hz and acoustic intensities between 90 and 120 dB. Expanding on its versatile applications, as PENG has illustrated its sensory functions in the human nervous system, recent efforts are focusing on employing PENG-based devices to simulate sensory integration processes. *Chen* et al. engineered a piezotronic graphene artificial synapse for neuromorphic computing applications [85]. Their approach leveraged P(VDF-TrFE)-based PENGs as pre-synaptic triggers to modulate an ion-gel gated graphene field effect transistor (FET) in the post-synapse (Fig. 3C). The system successfully mimicked several vital biophysical features found in human synapses. Strain-induced piezoelectric signals from PENGs could produce both excitatory postsynaptic current (EPSC) and inhibitory postsynaptic current (IPSC), which play a pivotal role in the formation and transmission of new action potentials in subsequent neurons. In addition, synaptic plasticity was effectively demonstrated: synaptic weights can be modulated directly by varying the strain pulses. Overall, PENG technology has showcased its versatility in mimicking tactile and auditory inputs for the human nervous system, and cutting-edge advancements in PENG-based neuromorphic computing are emerging to meet the growing demands for sensory integration and computation.

Existing literature has already applied PENG in neuromodulation therapeutics targeting both the central nervous system (CNS) and peripheral nervous system (PNS). In addition to harvesting energy from intrinsic body motions, PENGs could be powered by external sources, such as ultrasound, and deliver neurostimulations. This method is helpful for reaching deep brain targets when extending wires through the brain tissue may pose a potential hazard. *Hwang* et al. constructed a piezoelectric energy harvester to power a deep brain stimulator device (Fig. 3D) [86]. The harvester employed materials such as PIMNT, Au, and PET and was characterized by an open-circuit voltage of 11V and a short-circuit current of 285 μA. Once connected to a stimulation electrode implanted into the mouse primary motor cortex (M1), the PIMNT-based piezoelectric energy harvester succeeded in controlling mouse limb movement (Fig. 3E). *Lin* et al. engineered a self-powered wearable brain-machine interface (BMI) system capable of both monitoring and providing neurostimulations to mitigate epilepsy [86]. The architecture of this system featured a PZT-based PENG as its energy harvester, complemented by a PVDF-ZnO-based motion detector, a data-processing module, and a neurostimulator. Following the detection of epileptic-related seizures by the PVDF-ZnO-based motion detector, the neurostimulator, powered by daily motions through the PZT-PENG, delivered electrical pulses to counter epilepsy. *In vivo* experiments on mice with kainic acid-induced epilepsy, stimulation electrodes were implanted in the dentate gyrus of the hippocampus. The results demonstrated that the group of mice receiving the stimulation experienced epileptic seizures of significantly shorter duration (143.2 ± 8.9 s) compared to the control group (313.2 ± 71.9 s). These findings corroborate the system's efficacy in mitigating epileptic episodes. Alternatively, *Guan* et al. proposed a design that involves a probe acoustically driven by smartphones for intelligent and remote neurostimulation in epilepsy treatments (Fig. 3F) [88]. The probe featured a pedestal with the neurostimulation electrode, and a resonator encapsulated by a PDMS outer shell and an epoxy inner shell. It also integrated a PZT-based piezoelectric film and a signal modulation unit to harvest acoustic energy and receive acoustic commands. Mice, receiving kainic acid injections mimicking epileptic incidences, were implanted with a brain probe in the dentate gyrus of the hippocampus for *in vivo* experiments. In a head-fixed setup, mice were exposed to virtual reality stimuli while their limbs remained free to move on a floating ball. Following acoustically modulated neural stimulation, a significant decrease was observed in the frequency and amplitude of limb movements, thereby establishing the probe's efficacy in epilepsy management. These changes were tracked using AI-based animal monitoring software. In a separate setup where the mice were free to move, large ictal spikes were less frequent. Additionally, the power across various EEG frequency bands (delta, theta, beta, and gamma) showed a decrease in the group treated with acoustically driven neurostimulation. These results suggest effective mitigation of epileptic seizure activities, as evidenced by concurrent intracortical EEG recordings. This innovative brain probe can be remotely powered and modulated via smartphones, opening the door for medical professionals and AI systems to deliver precise, real-time stimulation therapies. *Zhang* et al. introduced a piezoelectric ultrasound-energy harvester (PUEH) designed for deep brain stimulation [87]. The device is engineered to incorporate a Sm-PMN-PT single crystal, which not only guarantees a safe operational range but also provides outstanding performance metrics. These include a piezoelectric coefficient (d33) of 4000 pC/N and an electromechanical coupling coefficient (k33) of 95 %. Additionally, the device boasts a relative permittivity (ε) of 13,000, an instantaneous output power of 1.1 W cm^{-2}, and an average charging power of 4270 ± 40 nW. *In vivo* experiments focused on the periaqueductal gray (PAG), a critical brain area for pain perception, showed that 1 MHz-ultrasound-induced neurostimulation led to analgesia in mice subjected to formalin-induced pain. Moreover, *Zhao* et al. developed a PZT/PVDF-based PENG system that could bidirectionally modulate synaptic neuroplasticity [156]. Powered by the mechanical force of a human pressing the device, the PENG generated high-frequency (HFS) or low-frequency (LFS) stimulations to induce long-term potentiation (LTP), in which synaptic connections were strengthened, or long-term depression (LTD), in which synaptic connections were weakened, respectively. These processes are crucial for the formation of memory and the facilitation of higher cognitive functions. During the *in vivo* experiments, stimulation electrodes powered by the PENG were implanted into the Schaffer Collateral of the Cornus Ammonis 3 (CA3) region while simultaneous recording was conducted in the Cornus Ammonis 1 (CA1) region of the mice hippocampus to capture the field excitatory postsynaptic potential (fEPSP). Applying either HFS at 100 Hz or LFS at 1 Hz resulted in either an increase or decrease in the slope of fEPSP, respectively. This demonstrated that both LTP and LTD were successfully induced respectively.

PENG-driven neurostimulations have also been broadly explored as modulatory therapeutic approaches for the peripheral nervous system. *Lewandowski* et al. presented an implantable, muscle-stimulated piezoelectric energy harvesting generator [157]. The piezoelectric stack energy harvester was surgically implanted between a bone and the muscle-tendon unit of a rabbit's quadriceps. This configuration drove tripolar spiral nerve cuff electrodes that delivered stimulations to motor neurons. This setup effectively demonstrated that the electrical power, converted from mechanical power generated by muscle contraction through the piezoelectric energy harvester, was adequate for stimulating the same motor neuron. The study thus laid a solid foundation for closed-loop neuromodulation techniques that efficiently harvest mechanical energy from rhythmic skeletal muscle movements to electrically stimulate adjacent motor neurons. *Zhang* et al. developed a PENG device capable of harvesting energy from the pulsations in carotid arteries for vagus nerve stimulation (VNS) [158]. Utilizing electrospinning technology, the researchers team employed a [P(VDF-TrFE)]/$BaTiO_3$ multilayer design for the PENG, achieving a maximum open-circuit voltage of 84 V and a short-circuit current of 1.32 μA. *In vivo* testing of this PENG-based VNS system on canine models confirmed its efficacy. Specifically, after the PENG gathered energy from the carotid artery pulses, subsequent stimulation deliveries to the vagus nerve led to a reduction in heart rate without any observable changes in systolic blood pressure. Additionally, the system featured a reed switch activated externally by a magnet, to release the stored electrical energy stored in the capacitor.

In addition, PENG and piezoelectric materials have been successfully employed in the realms of neural regeneration and neural repair. *Qian* et al. engineered a nanogenerator scaffold composed of ZnO-loaded polycaprolactone (PCL) to facilitate neural regeneration and motor recovery [159]. The ZnO/PCL PENG neural scaffold demonstrated exhibited high biocompatibility with Schwann cells (SC) *in vitro*, as evidenced by its elevated expression of key neuro-regenerative proteins, including nerve growth factor (NGF) and vascular endothelial growth factor (VEGF), compared to PCL alone. The system was subsequently tested in mice with severed sciatic nerves *in vivo*. Mice were implanted with either a ZnO/PCL scaffold, a PCL scaffold, or an autograft and underwent physical therapies (PT+) or not (PT-), resulting in 6 distinct experimental groups. Physical therapies through treadmill running were employed to deform the piezoelectric materials, thereby generating electricity. Mice implanted with the ZnO/PCL scaffold and subjected to physical therapies exhibited the highest expression levels of GFAP, a critical biomarker for distal glial cell viability, and β–III–tubulin (Tuj1), as well as optimal nerve conduction velocity (45.4 m/s) and significant improvements in motor recovery related to endplate muscle (gastrocnemius muscle) weight, morphology, and fiber type. Additionally, *Lee* et al. explored the use of electrospun fibrous PVDF-TrFE piezoelectric conduits in combination with SC for the purpose of spinal cord injury recovery [160]. A complete spinal cord transection model was created in rats, and circular PVDF-TrFE conduits were implanted between the rostral and caudal stumps, with fibers either being randomly or longitudinally aligned with the spinal cord. GFP-SCs, mixed with Matrigel, demonstrated robust viability within these conduits. In both conduit configurations, sensory axons were observed to regenerate at the caudal interface. However, SCs were more uniformly dispersed and constructed a denser tissue bridge in the longitudinally aligned conduit compared to the randomly aligned one.

Furthermore, piezoelectric materials have shown efficacy in guiding stem cell differentiation. *Zhang* et al. used piezoelectric biomaterials and nanotopographic substrates to stimulate the neural differentiation of rat bone marrow-derived mesenchymal stem cells (rbMSCs) [161]. The experimental design involved two kinds of piezoelectric PVDF materials with nanoscale stripe arrays of stripes depth and width of 200 nm (PVDF-200) or 500 nm (PVDF-500), and two corresponding non-piezoelectric PVC materials, PVC-200 and PVC-500. *In vitro* assays revealed that the neuron-specific marker Tuj-1 was more abundantly expressed in the PVDF group. It was also found that the more nano-topographic PVDF-200 setup generated a higher piezoelectric potential than PVDF-500, thereby leading to an increased number of Tuj-1 positive cells. This comprehensive set of results underscores the dual roles of piezoelectricity and nano-topography in neural differentiation. *Tai* et al. designed a piezoelectric nanofibrous scaffold constructed from P(VDF-TrFE) that could power electrical stimulation for inducing multi-phenotypic differentiation of neural stem cells into neuronal, oligodendrocytic, and astrocytic phenotypes [162]. Cells placed on either piezoelectric or piezo-inactivated scaffolds underwent different types of stimulation, specifically after thermal treatment. These conditions included electrical stimulation (ES), in which cells on piezo-inactivated scaffolds were directly electrified; mechanical stimulation (MS), where these cells were subjected to hydro-acoustic actuation through periodic vertical movements; and mechano-electrical stimulation (MES), where cells on piezoelectric scaffolds received both electrical and mechanical cues via hydro-acoustic actuation. Neuronal biomarkers, such as β-tubulin III (Tubb3), microtubule associated protein 2 (MAP2), and Eno2 were upregulated in the MES condition, alongside oligodendrocyte markers Olig1 and Cldn11, and astrocytic genes Aldh1l1 and Cspg4. The study further explored the morphological development of mouse neural stem cells (mNSC) and human neural stem cells (hNSC) under either static or hydro-acoustically actuated culture conditions. Under static conditions, neuronal differentiation predominated, as evidenced by the higher expression of neuronal biomarker NeuN. In contrast, hydro-acoustic actuation led to spatially differentiated cellular types, with neurons predominantly located in the upper layer and glial cells adjacent to the scaffold surface, marked by high expressions of astrocyte marker ALDH1L1 and oligodendrocyte marker O_4. In conclusion, a variety of PENG and piezoelectric materials have shown promising results in both neural regeneration and differentiation, primarily through electrical stimulation mechanisms.

3.3. Magnetoelastic nanogenerators (MEGs)

3.3.1. Working principles

Since its first report in 2021 [28], the field of magnetoelastic nanogenerators (MEGs) has received considerable attention due to its huge potential in both wearable and implantable bioelectronics [163]. MEGs primarily utilize the giant magnetoelastic effect of soft magnetic materials to generate electricity from biomechanical movements. An MEG comprises two key layers, including a magnetomechanical coupling (MC) layer, where changes in the magnetic field occur under mechanical stress, and a magnetic induction (MI) layer, where these magnetic field variations are picked up by MI layer to generate electricity through a wire coil.

In the MC layer, a soft magnetoelastic film is produced by combining porous polymers with nano-/micro-magnets. The film can deform in response to external stress. In the relaxed state, nano-/micro-magnets within the MEG align according to a wavy chain configuration. In the compressed state upon the application of external force, the aligned directions of the nano-/micro-magnets rotate, leading to a decrease in magnetic flux density (Fig. 4A). Such decrease at microscale causes the shape deformation resulting in magnetic particle-particle interactions. While at atomic scale, magnetic dipole-dipole interaction was also introduced by the mechanical stress. Soft magnetoelastic materials have demonstrated a much higher magnetoelastic coupling factor (6.77×10^{-8} T/Pa) than the rigid metal and metal alloy counterparts, serving as the foundation for their high-performance biomechanical-to-electrical energy conversion. MEGs have found applications in respiratory and cardiovascular monitoring, as well as human-computer interactions [28,90,91]. Importantly, MEGs offer high biocompatibility and intrinsic waterproofness, making them ideal for implantable biomedical systems.

3.3.2. Applications in self-powered neurostimulations

Wearable MEGs show great promise in mechanical energy harvesting [92]. By harnessing this energy, electrical stimulations can be directed to neural tissue. *Libanori* et al. constructed an MEG-based array capable of delivering electrical stimulations (ES) for cell reprogramming [89]. The MEG was assembled by mixing NdFeB nanoparticles with a liquid silicone rubber solution to form the MC layer, while the MI layer was formed using wound copper (Cu) wire coils. This intricate design exploits both the giant magnetoelastic effect and MI, thereby transforming mechanical deformations in the MC layer into electricity. An air pressure-activated MEG ES platform hosting fibroblasts transduced with neuronal BAM TFs (Brn2, Ascl1, and Mytl1) was developed. This platform is capable of generating a current of up to 10.5 mA and a voltage of

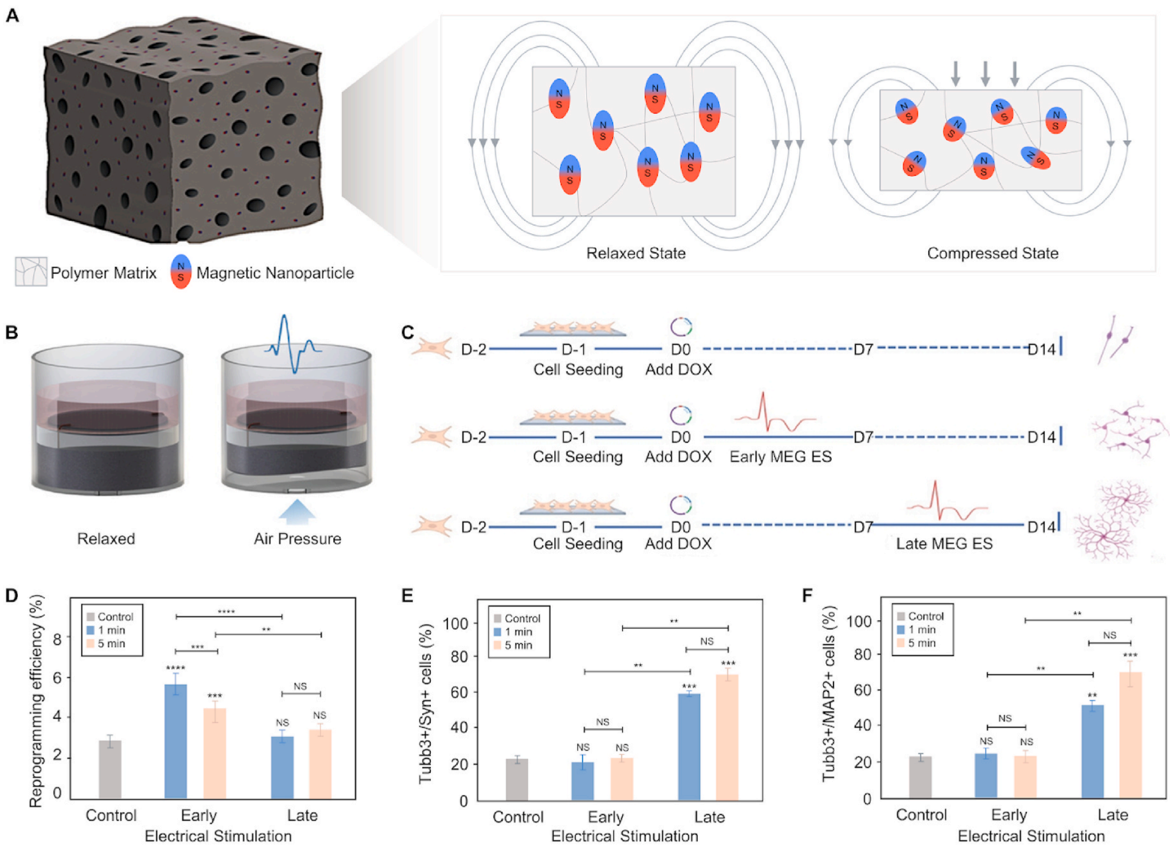

Fig. 4. Working principles of MEGs and their applications in neurostimulation. (A) Clarifies the fundamental working principle of MEG by providing a microscopic view of the changes in magnetic dipole orientation and magnetic flux density when transitioning between relaxed and compressed states [103]. (A) Reproduced with permission. Copyright 2022, Wiley-VCH. (B) Offers schematics of MEG-driven electrical stimulation, where deformation induced by air pressure in the MEG leads to the generation of electrical energy. (B) Reproduced with permission. Copyright 2022, Wiley-VCH. (C) Details the experimental design for administering MEG-facilitated electrical stimulation at varying intervals as part of cell programming (D: day; Dox: doxycycline). (C) Reproduced with permission. Copyright 2022, Wiley-VCH. (D) Showcases the efficiency of cell reprogramming from fibroblasts to neuronal types under different ES settings, as quantified by the number of Tubulin Beta 3 Class III+ (Tubb3+) cells identified on day 14. $^{**}p \le 0.01$, $^{***}p \le 0.001$, $^{****}p \le 0.0001$, NS = not significant. (D) Reproduced with permission. Copyright 2022, Wiley-VCH. (E) Assesses mature neuron cells under dissimilar ES conditions, gauged by the number of Tubb3+ and Synapsin + cells identified on day 14 relative to Tubb3+ cells. $^{**}p \le 0.01$, $^{***}p \le 0.001$, $^{****}p \le 0.0001$, NS = not significant. (E) Reproduced with permission. Copyright 2022, Wiley-VCH. (F) Evaluates mature neuron cells under divergent ES conditions, as determined by the number of Tubb3+ and Microtubule Associated Protein 2 (MAP2+) cells identified on day 14 relative to Tubb3+ cells. $^{**}p \le 0.01$, $^{***}p \le 0.001$, $^{****}p \le 0.0001$, NS = not significant. (F) Reproduced with permission. Copyright 2022, Wiley-VCH.

9.5 mV under air pressure ranges between 100 and 400 kPa (Fig. 4B). The researchers also explored the timing of fibroblast stimulation. Stimulations were either applied during the first 7 days of a 14-day period (early stage), or during the last 7 days of the 14-day period (late stage), or not applied at all as a control (Fig. 4C). Transduced fibroblasts were exposed to electrical pulses at a frequency of 1 Hz and a current of 50 nA for either 1 or 5 min. The results indicated that early-stage stimulation significantly enhanced the reprogramming efficiency of fibroblasts, as evidenced by the higher percentage of induced neurons expressing neuronal β-tubulin III (Tubb3+), compared to late-stage stimulations. Moreover, the duration of the stimulation was found to play an important role in this context (Fig. 4D). Conversely, the maturation of neurons, as denoted by the expression of mature neuronal markers such as Synapsin (Syn) and microtubule associated protein 2 (MAP2), was predominantly observed during late-stage stimulations (Fig. 4E and F). Overall, the study demonstrates that temporally coordinated ES, powered by MEGs, can effectively reprogram and mature fibroblasts into neurons. Although this is the only study to date that employs MEG as an energy source for neurostimulation, it suggests that MEGs hold significant promise for advancing future neurostimulation systems, much like other mechanical energy-harvesting nanogenerators such as TENGs and PENGs.

3.4. Biofuel cells

3.4.1. Working principles

Biofuel cells (BFCs) can convert biochemical energy directly into electrical energy through ongoing redox reactions in the biofluids. These cells can generate electricity through charge transfers in the ubiquitously occurring glucose oxidation reactions. In a BFC, the bioanode incorporates oxidizing biocatalysts to expedite the oxidation of biofuels, such as glucose, and releases electrons. These electrons migrate toward the biocathode which uses reducing biocatalysts to catalyze the reduction of Oxygen to water (Fig. 5A). The cumulative flow of electrons from the bioanode to the biocathode generates an electrical current for self-powered neurostimulation. BFCs can be generally categorized into two major classes: microbial fuel cells (MFCs), which include microorganisms, proton exchange membranes (PEM), and electron acceptors, and enzyme fuel cells (EFCs), which use enzymes as the biocatalysts for the redox reaction [164]. Due to their renewability and sustainability, BFCs offer significant potential as power sources for a variety of implantable and wearable bioelectronic devices [165–167].

3.4.2. Applications in self-powered neurostimulations

Biofuel cells have been predominantly used to harvest biochemical

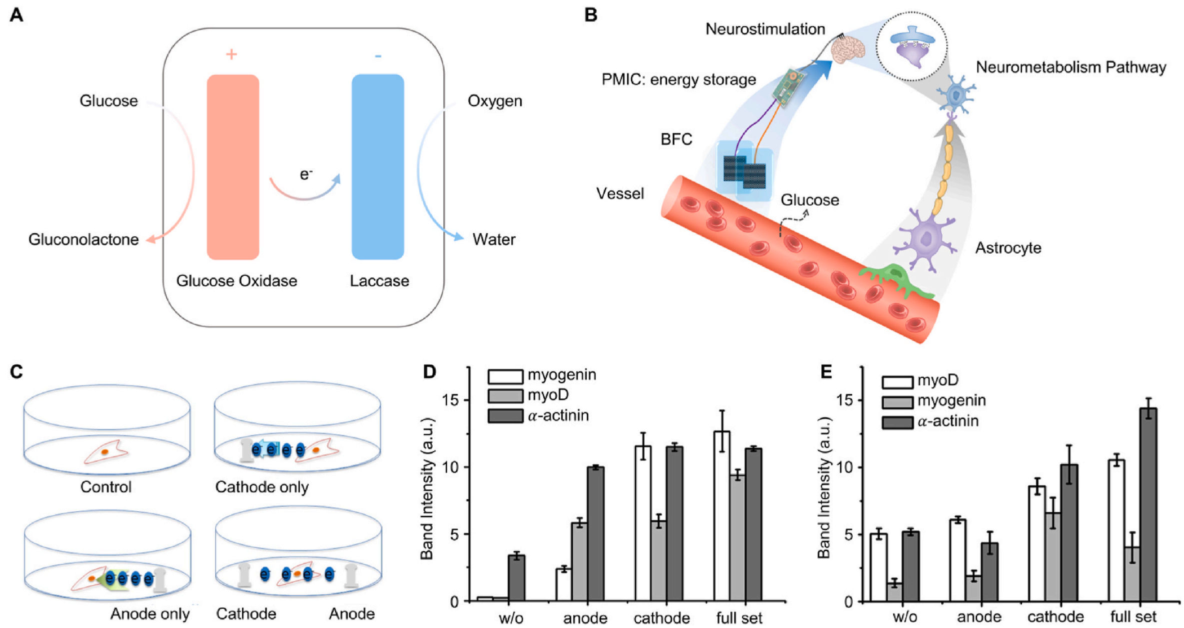

Fig. 5. Operational mechanisms of biofuel cells and their applications in neurostimulation. (A) An overview depicting the biochemical process in a biofuel cell, using glucose oxidation as a model reaction. (B) Graphic outline of the conversion of chemical energy, generated by glucose, into electrical energy suitable for neurostimulation. This is achieved via a biofuel cell (BFC), which stores the energy in a Power Management Integrated Circuit (PMIC) [90]. (B) Reproduced with permission. Copyright 2020, Elsevier. (C) Visual representation of four unique experimental designs for the Enzymatic Biofuel Cell (EBFC), aimed to stimulate muscle precursor cells (C2C12) in an *in vitro* environment [92]. (C) Reproduced with permission. Copyright 2015, Elsevier. D) RT-PCR results indicating gene expression levels associated with myogenesis in C2C12 cells (myogenin, myoD, **α**-actinin) across the multiple experimental conditions [92]. (D) Reproduced with permission. Copyright 2015, Elsevier. (E) RT-PCR results highlighting gene expression levels linked to myogenesis of C2C12 cells (myogenin, myoD, **α**-actinin) across the four experimental setups, when housed within a PCL-gelatin based nanofibrous scaffold [92]. (E) Reproduced with permission. Copyright 2015, Elsevier.

energy within biological systems to power neurostimulators. *Lee* et al. developed an enzymatic BFC capable of supplying power to a brain stimulator implanted in pigeons [168]. They integrated a power management integrated circuit (PMIC) to store energy harvested from the BFC, which could subsequently be discharged wirelessly to deliver neurostimulation (Fig. 5B). The BFC harvested energy from glucose oxidation and oxygen reduction, using multi-walled carbon nanotube (MWCNT)-based biocathodes and bioanodes, and achieved a power output of 0.12 mW *in vitro* and 0.08 mW *in vivo*. In their *in vivo* experiments, the pigeon's brain stimulator was charged and discharged cyclically via wireless commands and was demonstrated to successfully alter the bird's behavioral responses. *Andoralov* et al. developed an EFC that can interface with living neurons [169]. The EFC comprised cellobiose dehydrogenase (CDH) as the glucose-oxidizing enzyme and bilirubin oxidase (BOx) as the oxygen-reducing enzyme, with CtCDH/AuNP/Au as the bioanode and MvBOx/AuNP/Au as the biocathode. *In vitro* testing in cerebrospinal fluid (CSF) environments showed that the EFC had an open-circuit voltage of 0.57 ± 0.01 V and a maximum power density of 7 μW cm^{-2}. The system also exhibited strong stability, maintaining 85 % of its initial voltage after 2 h. *In vivo* tests in mouse brains revealed a maximum power density of 2 μW cm^{-2}. Through electrochemical impedance spectroscopy, they observed an inductive loop only when the EFC was implanted in the mouse brain, a phenomenon not seen in buffer or CSF tests.

In addition, BFCs demonstrated their capacities in neural regeneration and repair. *Lee* et al. created an enzymatic BFC (EBFC) with the purpose of delivering electrical stimulation to promote the growth of muscle precursor cells, thereby aiding in the repair of neuromuscular functions [170]. They used glucose oxidase from Aspergillus niger as the anodic catalyst and BOx from Myrothecium verrucaria as the cathodic catalyst. They then performed *in vitro* tests on C2C12 muscle precursor cells to evaluate their myogenic expression. These cells were exposed to different electrical stimulations: no electrodes (as a control), an anode only, a cathode only, or both an anode and cathode (Fig. 5C). RT-PCR assays were conducted to measure mRNA expression levels of specific myoblast markers—such as myogenin, myoD, and α-actinin—which were used to assess myogenic activity. Remarkably, all groups exposed to EBFC electrical stimulation had significantly higher levels of myogenin and myoD compared to the control group. However, cells receiving both cathodic and full-set stimulation showed a notably greater expression of myogenin compared to those stimulated by the anode alone (Fig. 5D). The researchers extended their study to explore the expression behaviors of C2C12 cells within a PCL-gelatin based nanofibrous scaffold in an EBFC environment. Here again, cathodic and full-set stimulations were far more effective in enhancing expression levels than anodic stimulation alone (Fig. 5E). Separately, *Arslan* et al. made an innovative contribution by demonstrating the generation of bioelectricity using human neuronal-like cell SH-SY5Y cells in single-chamber air cathode biofuel cells [171]. The BFCs were capable of powering LEDs with a maximum power density of 12 mW m^{-2} and a current density of 0.013 mA cm^{-2}. This groundbreaking work suggests significant biomedical applications, as such BFCs could potentially serve as reliable power sources for future neurostimulators. Nevertheless, it's crucial to further investigate the electrophysiological behaviors and network responses of these biofuel neurons to ensure both functional performance and biocompatibility.

4. Prospect and conclusion

In this review, we have meticulously compiled studies focused on biomaterials and bioelectronics for self-powered neurostimulation with electrical stimulation. Continuously acquiring neural signals and delivering therapeutics serve as the bedrock for the prevention, diagnosis, and treatment of neurological diseases [172–175]. As the global population ages, the urgency for state-of-the-art neuromonitoring and therapeutic devices escalates, thereby amplifying the need for innovative research, product development, and streamlined commercialization processes [176,177]. Remote and wireless technologies within the neuroengineering sphere significantly bolster both the preventive and diagnostic measures for patients, in addition to aiding medical professionals in precise, timely decision-making [178–180].

While the surging demand for neuroengineering bioelectronics signals promising avenues, numerous challenges and limitations also pervade the research and methodologies examined in this paper. Neurostimulation requires long-term implantation and precise delivery of electrical stimulation. Proceeding issues of reduction of immune response, chemical and physical safety exists for any type of implantable device designed for delivering ES. Beyond that, there are some specific issues of self-powered devices, as demonstrated in Fig. 6A. Chief among these is biocompatibility, a critical factor for the successful amalgamation of biomaterials and bioelectronics in self-powered neural systems [181–183]. Adverse reactions between implanted devices and biological systems can trigger inflammation, thereby jeopardizing both the device's effectiveness and the integrity of surrounding tissues [184–186]. Some piezoelectric materials, such as PZT, present toxicity to the human body, which underlines the necessity for prioritizing biocompatible, non-toxic materials in future investigations [187–189]. Longevity and reliability are essential attributes for the practical application of self-powered devices in neural engineering [190–192]. For these devices to be clinically viable, they must demonstrate resilience to physiological conditions, such as the corrosive impact of bodily fluids and varying mechanical pressures [193]. This is especially relevant for devices that employ TENG technology, where material degradation could significantly hinder the performance [194–196]. Research agendas should target the fortification of device durability through novel designs, material choices, and defensive coatings. Optimal and consistent output performance remains a critical objective. Variability in energy harvesting and electrical output could compromise the effectiveness of neurostimulation and recording procedures [197,198]. Addressing the high internal impedance in TENG and PENG devices is paramount, especially given the preference for current-driven stimulation in neuroengineering applications [188,197,198]. Material selection remains a multifaceted issue, closely related to biocompatibility, durability, and output performance [53,199]. It should consider mechanical properties, electrical conductivity, biodegradability, and interaction with biological tissues. For methods like piezoelectricity and BFCs, material selection is constrained due to subtle principal requirements [49,200–202].

The promise of integrating biomaterials and bioelectronics offers a tantalizing glimpse into the future of self-powered neural engineering. However, resolving challenges associated with biocompatibility, durability, output performance, and material selection is indispensable for harnessing the full potential of these emerging technologies. MEG technology, as previously discussed, presents a promising solution to many of these challenges, offering favorable biocompatibility, lower internal impedance, and an expansive material selection range [90]. Moreover, due to its low inner impedance, MEG can produce a high current output compared to other methods, which lowered the total amount of energy required by the system and further ensure the safety of the device, especially in the scenarios like deep-brain stimulation. Harnessing the expertise from diverse fields like materials science, bioengineering, and neuroscience will be key in crafting groundbreaking solutions and broadening the scope of neural engineering.

Considering the far-reaching impacts of the COVID-19 pandemic beyond the healthcare sector, the relevance of self-powered neuroengineering becomes even more poignant. The pandemic has exacerbated the prevalence of lifestyle-related diseases like chronic pain and depression while amplifying investments in neurological disorder research [203,204]. Technological advancements, particularly remote control and closed-loop systems, represent groundbreaking developments in self-powered neural engineering [205–209]. These platforms allow real-time data capture and adjustments, thus enabling more adaptive and precise therapeutic interventions [210–213]. Furthermore, the rise of an aging demographic and growing consumer appetite for non-invasive surgical solutions act as catalysts for industry expansion. Future innovations should aim to miniaturize devices while maximizing flexibility and biocompatibility, thereby mitigating associated risks. Material innovation and advanced fabrication techniques are pivotal for actualizing highly effective neural engineering solutions [214–217]. The conceptualization of a whole-body network provides a holistic framework for neural engineering, enabling interconnected self-powered devices to coordinate interventions [205,218]. Creating robust communication protocols and interoperability standards is essential for operationalizing these intricate networks, paving the way for comprehensive healthcare management [219–224]. Personalized healthcare remains at the vanguard of medical advancements, with self-powered neural engineering poised to play an integral role [138,225,226]. The interplay between advanced sensing technologies and data analytics can potentially revolutionize personalized healthcare, allowing for tailored interventions specific to individual physiological and pathological states [227,228].

Given these evolving dynamics and optimistic trajectories for both research and market demand, neuromodulation is experiencing a marked surge in interest, a trajectory expected to continue. As illustrated in Fig. 6B, the global neuromodulation devices market stood at a valuation of USD 3.19 billion in 2021. It is projected to grow at a compound annual growth rate (CAGR) of 9.5 % from 2022 to 2030 [229]. A report

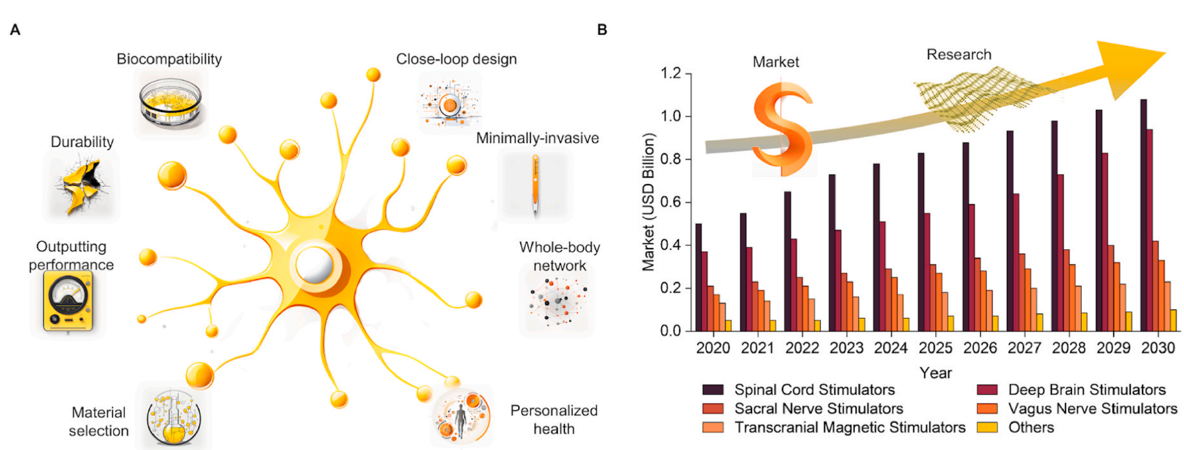

Fig. 6. Prospective developments in bioelectronics for self-powered neurostimulation. (A) Underscores not only the existing enthusiasm but also various underexplored areas and opportunities within the self-powered neurostimulation landscape, thereby delineating potential pathways for future device innovation. (B) Showcases the accelerating trajectory of progress in the domain of self-powered devices, as evidenced by increased attention and capital influx in both academic and commercial sectors related to neuromodulation.

by the American Academy of Neurology states that nearly 1 million people in the U.S. are affected by Parkinson's disease, with an additional 60,000 new cases diagnosed each year. Furthermore, data published by the Hearing Health Foundation in 2020 in the U.S. alone, 48 million individuals—and an estimated 477 million people globally—suffer from auditory conditions like deafness and tinnitus [230]. According to a factsheet by the World Health Organization, approximately 50 million people globally suffer from epilepsy, with close to 2.4 million new cases emerging each year [231]. As a result of the escalating prevalence of neurological conditions, demand for neuromodulation devices is poised to expand throughout the period under consideration [232–238]. The advent of technologies such as the Internet of Things (IoT) and personalized healthcare approaches particularly amplify the growing necessity for self-sustaining bioelectronics in the realms of neuromodulation and other neuroengineering applications [239–246].

In conclusion, the prospects for self-powered neural engineering are myriad, with focus areas including closed-loop systems, minimally invasive technologies, body area networks, and personalized healthcare solutions. The fusion of multidisciplinary research and technological innovation holds the key to surmounting existing challenges and unlocking untapped potential, ultimately contributing to human healthcare, and elevating the quality of our life.

CRediT authorship contribution statement

Jinlong Li: Investigation. **Ziyuan Che:** Investigation. **Xiao Wan:** Writing - review & editing. **Farid Manshaii:** Writing - review & editing. **Jing Xu:** Writing - review & editing. **Jun Chen:** Conceptualization, Investigation, Supervision, Writing - review & editing.

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.

Data availability

Data will be made available on request.

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