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Advanced 3D printing-based triboelectric nanogenerator for mechanical energy harvesting and self-powered sensing

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Triboelectric nanogenerator (TENG) is an innovative technology that it has sparked a revolution in distributed energy supply and self-powered system. Integration of advanced TENG with burgeoning 3D printing (3DP) technologies fosters the emergence of 3DP-based TENGs. It will inevitably promote the rapid development and widespread applications of next-generation portable electronics and multi-faceted artificial intelligence. However, due to the different subject field between researchers specializing in TENG and those good at 3DP, they are not always a perfect combination. It is rather difficult to achieve with both excellent electrical properties and outstanding practical performances. For that, a review is presented more systematic and comprehensive of 3DP-based TENGs for the first time. In which the quantitatively statistics and correlation data of research progress are given, such as publications, 3DP technologies and materials, structure designs and functionalities, working modes and mechanisms, output performances, unique advantages, potential technical challenges and promising application fields that can impede their sizable production and applications are discussed. It is hoped that this review will not only deepen the intersection and amalgamation between 3DP and TENGs, but also push forward more in-depth research and applications of future TENGs.

Keywords: 3D printing-based; Triboelectric nanogenerator; Energy harvesting; Self-powered systems; Distributed energy

Introduction

Nowadays, the rapid advancement of multifunctional electronic devices and energy technologies are changing every aspect of our daily life and the way of working, as it confirmed that the electricity and its way of supply have played the most critical role in the history of the technology revolution. Starting from the electromagnetic induction discovered by Faraday in august 1831 [1] mechanical energy was converted into electric power that can be generated by the burning of fossil fuels, wind, hydropower and nuclear fission, which has marked the mankind has entered an entirely new age of electricity. And invented solar cells based on photovoltaic effect is a kind of green energy that has succeeded in converting sunlight directly into electrical energy [2] which is widely used in the fields of energy storage and aerospace industry. In a way, with the development and utilize of electricity, this has symbolized the progressive degree of human society and the developed level of science and technology. However, with the rapid consumption of finite fossil fuels as well as people fully have woken to the importance of environmental protection, in which the traditional centralized and ordered energy supply patterns based on power plants are incompatible with the present development of distributed energy and

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portability electronic devices [3]. Besides, a comprehensive advantage of light weight, flexible, miniaturization and portability is needed. As the information age of applied 5G and 6G is coming, billions of things must be connected with various sensors for large data, artificial intelligence, face recognition, block chain, and many other new technology development and application [4]. These rapid advance mobile, individual, randomly, massively of the distributed and portability electronic devices also require the corresponding matched of energy supply pattern. In fact, it turns out that the unreasonable way of energy supply patterns and energy structure lead to the current development dilemma, i.e. one of the more prominent of them is frequent charging and its slow process [4]. Portable energy storage technologies are the most efficient solutions, that may be to provide a reasonable way for the above predicaments. For portable power sources, the most effective way now is to utilize the small scale, high energy density and rechargeable energy storage devices, which mainly include conventional solid-state battery, electrochemical capacitor, ultracapacitor and lithium ion battery, etc. However, with the inherent shortcomings of frequent charging, limited storage capacity, short service lifetime, certain safe hidden trouble, and serious environmental hazards, above all the electricity storage of these power sources still draws from conventional supply patterns. So that, these are also not fundamental solution for future distributed and portability electronic devices. Furthermore, from long-term development objective and environmental perspective, power acquisition directly from our natural environment is the ideal choice for future energy supply [5,6]. This idea was first proposed by Z.L. Wang in 2006 that is invented nanogenerator (NG), this technology is now well received and recognized worldwide [7,8]. Though the existing mature energy technologies show that have better performances for generate electricity, yet they are generally excessive reliance on external factors and supporting conditions, such as abundant sunshine, the proper temperature range and active catalyst, making them difficult to be effectively utilized on the energy supply of distributed and portable patterns.

With continual advances in energy nanotechnology, triboelectric nanogenerator (TENG, by Z.L. Wang discovered in 2012) as a revolutionary of mechanical energy harvesting technology stands out from the rest with its own advantages and stand-out features [9,10]. It makes possible to expediently recycle and converts mechanical energy into electrical energy by the coupling effect between the contact electrification and electrostatic induction, which is through the periodic contact-separation between two triboelectric materials with different abilities of gaining or losing electrical charges. This new approach to harvest mechanical energy can high-efficiency energy conversion low-frequency mechanical energies from our living environment, and is capable of supplying electricity for distributed and portability applications [11–14]. TENGs have received increasing interest among researchers in recent years and have been regarded as effective technological means to harvest mechanical energy and self-powered sensing, which have broad application prospects in distributed and portable energy technology, such as motion tracking, physiological monitoring, medical rehabilitation, intelligent humanoid robot and human–computer interaction [12–20]. Particularly, when advanced TENGs in combination with emerging 3D printing [21] (3DP, or umbrella term additive manufacturing technology gained popularity in the 2000s) contributes to the emergence of a series of revolutionary mechanical energy harvesting devices, i.e., termed 3DP-based TENGs, which make full use of their respective merits and will inevitably promote the rapid development of new era energy harvesting technology and self-powered sensing. This technical approach was first proposed by Z.L. Wang in 2018, while report a practical, ultraflexible and three-dimensional TENG that is capable of driving conventional electronics by harvesting biomechanical energy [22]. It is now well received worldwide and 3DP has been utilized to fabricate TENGs [23]. Fig. 1 demonstrates several kinds of 3DP-based TENGs as well as their structure and a more diverse set of real application, such as the motion of the human, vibration energy, wind energy and blue energy from the natural environment. It can be found that portable power socrce and self-powered sensing are their two main applications. It is usually used for distributed energy supply and active sensing in many aspect such as action, motion, pressure, tactile, healthcare, safe guarding, information acquisition, signals transmit of big data and more powerful human–machine interacting. The number of published research papers about 3DP-based TENGs from the top-ranked countries are demonstrated in Fig. 1b. The overall situation of publications about 3DP-based TENGs was investigated and annual number were statistical analysed, as shown in Fig. 1c. Although related researches are just starting, according to the latest published resultes, it is shown that 3DP-based TENGs studies range is wide, applied foreground is hopeful, simple structure, ease of operation and integration, huge potential of research. Due to the differences of mechanical energy in the environment, so have to optimizing design and select suitable process for the fabricated 3DP-based TENGs to be applicable to real-world applications. As technology develops, it can be believed that 3DP-based TENGs will bring new field of research and more possibilities for next-generation low-power electronics, and lead the way of people's life toward a more intelligent and portable developing direction in the near future.

As technology advances and more prototypes of 3DP-based TENG have been largely fabricated and extensively reported, several fundamental issues must finally confront together, such as current research status and practicability, 3DP technologies classification and characteristics, 3DP materials selection and preparation, integration methodology and structural design, working mechanisms, electrical properties improvement methods and promising application occasions, etc., at the moment these are still not comprehensively and systematically overviewed. And in particular, the basic knowledge of 3DP and the approaches to efficient integration TENG energy harvesting technologies with 3DP are also not roundly and comparatively summarized. These urgent issues have greatly bound the development of commercialization and industrialization of 3DP-based TENG, which results in a large gap between the best current devices and real applications.

Herein, we cover the recent progress of 3DP-based TENGs for both energy harvesting and self-powered sensing in the natural environment, discuss the major research challenges and also point out a clear direction for future development. We expect

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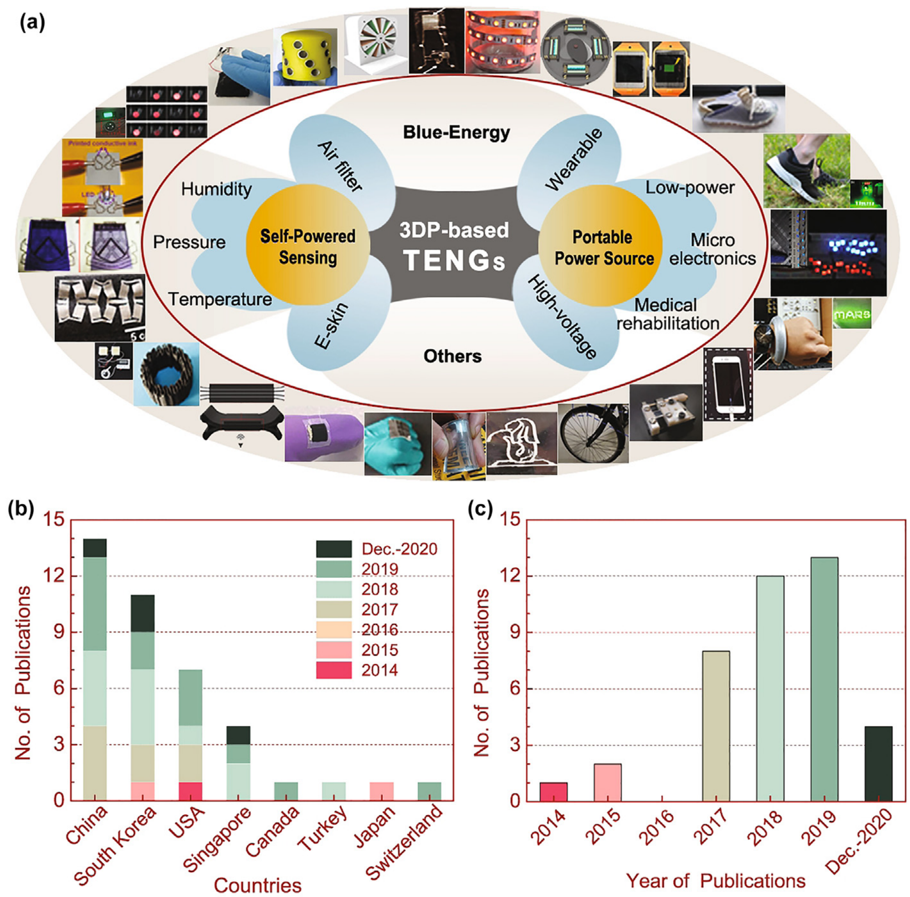

FIG. 1

Application demonstrations and the current research status of 3DP-based TENGs in different fields. (a) Application demonstrations of 3DP-based TENGs in different fields [16,24–58]. (b) The publications of 3DP-based TENGs from the top-ranked countries [16,24–58,69–76]. (c) The publications of 3DP-based TENGs in recent years [16,24–58,69–76].

this review article to greatly benefit the related developers and research teams. Our objectives not only gave a summary report for the research situation and applications, but also more importantly is to provide a reference for future research.

3DP technologies for TENGs

Clearly, TENG endow 3DP technologies with super-complex structure design carrier for their applications, while 3DP provide an integrated manufacturing platform for its research and development [24–28]. 3DP is a process of making three dimensional solid objects from a digital file, will be deeply affected the industrialization process. It is also considered a revolutionary and greatly attractive process for the fabrication of energy devices. With 3DP become more widely used in all walks of life, the resulting impact on the TENG applications will depend on how to use this technology. The process of 3DP is as follow: materials selection, model design, parameters setting, code generation, operating 3DP [29]. The SolidWorks software and Cura tools were used for 3D design and slicing, respectively. The code was generated and loaded during the printing process maintaining appropriate ink materials thickness, printing polymers spaces, contact frequencies, forces and printing directions.

Classification and features of 3DP technologies for TENGs

Fig. 2 summarily demonstrates several commonly used 3DP technologies for the fabrication of 3DP-based TENGs, including some of the flexible and rigid TENG structures. Several mature kinds of 3DP technologies are used for fabrication of TENG units as well as their printing process and a more diverse set of 3D printed structure are shown in the Fig. 2, such as filament, film, array, lattice, layered and 3D-network structure and so on [30–43,22]. As can be seen in the figure, four types of 3DP technology are mainly used for the fabrication and research of TENGs: fused deposition modeling (FDM) [30,31,32,34,35], direct ink writing (DIW) [33,36,39,40], stereo lithography appearance (SLA) [22] and selective laser sintering (SLS) [37,38,41,42], while some other 3DP technologies (typically lithography and coating) [43] are seldom used. Practice show that FDM technology suits to fabrication of the filament [30] film [31,32] and array structure [34,35] with thinner and tiny structures very well, as shown in Fig. 2a, b, c, e and f. It is very remarkable in technique-economic effect and in rapid prototyping technology is widely applied in industry. This particular aspect was well suited for the design and development of new triboelectric materials because of its lower

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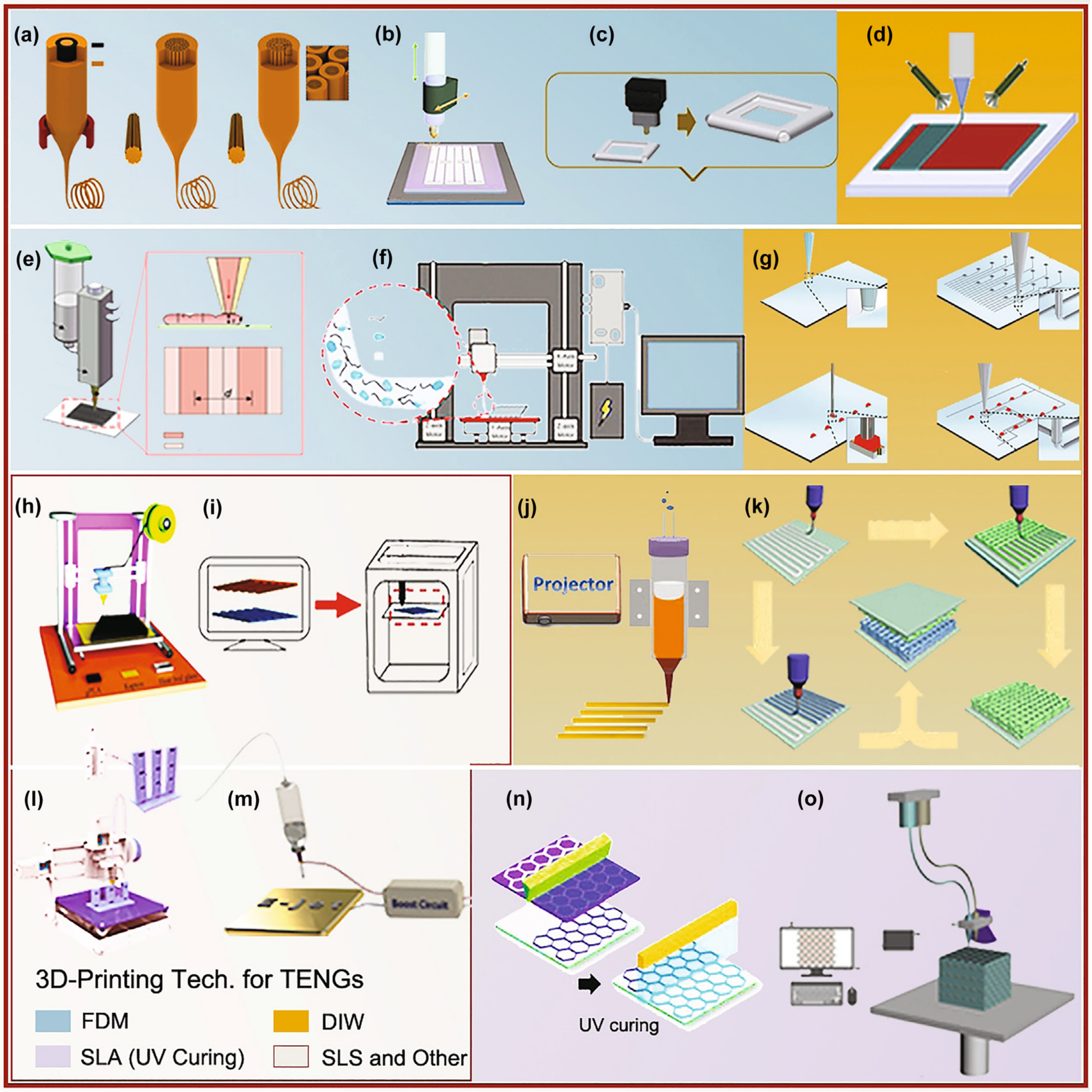

FIG. 2

Schematic illustrations and structure demonstrations of 3DP-based TENGs by various 3DP technologies. (a), (b), (c), (e) and (f) Schematic illustrations and structure demonstrations of 3DP-based TENGs by FDM technology [30,31,32,34,35]. (d), (g), (j) and (k) Schematic illustrations and structure demonstrations of 3DP-based TENGs by DIW technology [33,36,39,40]. (h), (i), (l) and (m) Schematic illustrations and structure demonstrations of 3DP-based TENGs by SLS and other technologies [37,38,41,42]. (n) and (o) Schematic illustrations and structure demonstrations of 3DP-based TENGs by SLA technology [43,22].

expense of equipment and lower consumption of materials. Moreover, DIW technology refers to a printing method based on extrusion through a nozzle under pressure, deposited along digitally defined paths to fabricate 3DP-based TENGs of layer-by-layer, as shown in Fig. 2d, g, j and k. That is an extrusion-based and heavily for the structures of meso-micro scales, such as to fabrication of the 2D lattice structure [33,36,39], multilayered geometries [40] with new triboelectric materials of different dopings, these are owes a good deal to the liquid-phase "ink" materials. Furthermore, SLA technology is a form of 3DP technology used for diverse variety of TENGs parts in a layer by layer fashion using photochemical processes by which light causes chemical monomers and oligomers to cross-link together to form polymers, as shown in Fig. 2n and o, suitable for processing flexible products which planar patterning, tridimensional, high-precision and shape more complex as well as in many other applications [43,22]. While this method is fast and can almost fabricate any structure units, but would be high costs and building up an additional supporting structure is needed. In addition, SLS technology uses a laser as the power source to sinter powdered material (typically nylon or polyamide), aiming the laser automatically at points in space defined by a 3D model, binding the material together to create a customized solid structure. 3DP-based TENG units were fabracited by SLS and other technologies, as shown in Fig. 2h, i, l and m. Practice also shows that it is fastest additive manufacturing process and efficient for the complex structures of 3D parts, such as to fabrication of the 3D lattice structure [37] complex geometrie [38] multilayered nesting [41,42]. In which the materials selection of SLS is wide enough for batch production, as well as the printed parts possesses excellent combined properties of high strength, stiffness, durability and good chemical resistance. In contrast with SLA, which most

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often require special support structures to fabricate overhanging designs, SLS does not need a separate feeder for support material because the part being constructed is surrounded by unsintered powder at all times, but the preheating and melting process are still an indispensable. Meanwhile, a list of data statistics are obtained, it is found that researchers were given priority to FDM approache for the fabrication of 3DP-based TENGs, where the percentage reached 67.5% of all, as shown in Fig. 3a. Other approaches are ranked as follows: SLA (10%), SLS (10%), DIW (7.5%) and other (5%). In a word, the biggest challenges of any 3DP-based TENGs is how to systematic integrate electrode layer and functional triboelectric materials, optimize electrical output performance, improve practical application capacity. So that, we should try different 3DP technologies and novel process to resolve these problems, as well as through optimizing process parameters to deal with the requirements of practical application.

Selection and optimization of 3DP materials for TENGs

TENGs had been invented since 2012 and has been widely utilized in modern self-powered systems to harvesting mechanical energy into electricity [44]. Enabled further by the invention of 3DP-based TENGs in 2018 [45,46]. Of which the selection and optimization of triboelectric materials for TENGs is crucial for the output performance and practicability by a energy harvesting device. We recently introduced a universal standard method to quantify the triboelectric series for a wide range of polymers, a fundamental materials property of quantitative triboelectrification is established by the way of normalized triboelectric charge density (TECD) [47]. This first quantitative triboelectric series will be a textbook standard for implementing the application of TENGs for energy harvesting and self-powered sensing. The methodology established will be extended to all conventional materials, such as ceramics, semiconductors, and polymers. Based on this, the different triboelectricm materials had been extensively studied and evaluated as well as is used for fabrication of 3DP-based TENGs [32,35,36,41,43]. According to the different production processes of 3DP, the functional triboelectric materials used for 3DP could be divided into typical liquid-phase and solid-phase, and it must be with reinforced triboelectric additives as auxiliary material at times. The existing researches have confirmed that the triboelectric series is widely used to choose appropriate triboelectric materials, and is considered to enhance the triboelectrification effect of 3DP materials. For solid-phase 3DP materials, some materials are appropriate the supporting structure in TENGs, such as crylonitrile butadiene styrene (ABS), polyethylene (PE) and polylactic acid (PLA). While others are more functional of triboelectric layer, such as nylon (PA), polyethylene glycol (PEG) polytetrafluoroethylene (PTFE). Those solid-phase materials generally suitable for the production of rigid 3DP-based TENGs. For liquid-phase 3DP materials, including polydimethylsiloxane (PDMS), poly (vinylidene fluoride) (PVDF), light-cure composite resin and hydrogel, etc., which is mainly used for fabricating the flexible and stretchable 3DP-based TENGs. Based on the published references statistics from 2012 to Dec. 2020, the use of triboelectric materials have satisfactory results in 3DP.The percentage of 3DP materials (into two categories: Solid-phase and Liquid-phase) for fabrication of TENGs, as shown inFig. 3b, them reached 62.5% and 37.5%, respectively. Overall, 3DP-based TENGs were fabricated through selection and modifying of 3DP materials which the stucture optimization and performance improvement were elementarily realized in relation to different usage scenarios.

Structure design of 3DP-based TENGs

Structural optimization of TENGs have especially important meaning for the output performance significantly enhancement, is the foundation that improve efficiency of energy harvesting, and the structure design is pivotal to a successful fabrication and application as well. More often than not, the fabrication process of 3DP-based TENGs is as follows: the 3D model is modeled by the Solidworks and 3D Max software, code generation, slicing, so as to guide the printer to print layer by layer. To this end, the structure design of 3DP-based TENGs had been widely researched with different structures in the published literature.

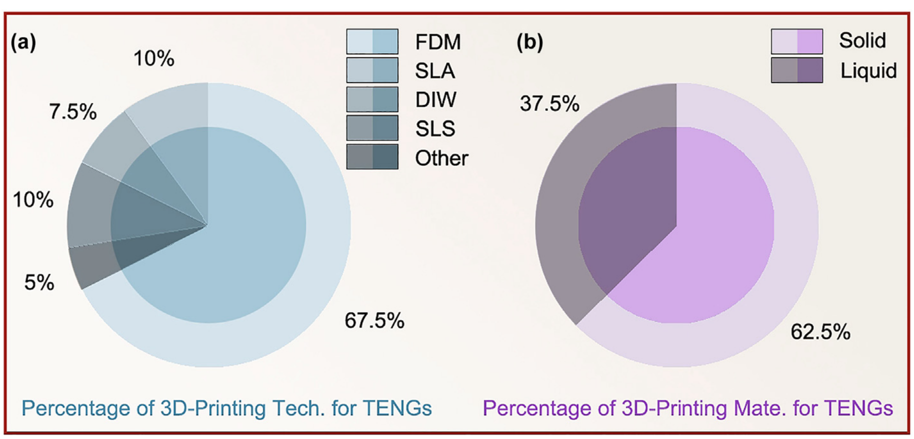

FIG. 3

The percentage and characteristics of 3DP technologies and materials on the fabrication process of TENGs. (a) The percentage distribution of 3DP technologies (into five categories: FDM, SLA, DIW, SLS and Other) on the fabrication process of TENGs. (b) The percentage distribution of 3DP materials (into two categories: Solid and Liquid) on the fabrication process of TENGs.

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Nevertheless, the applications of 3DP-based TENG in energy harvesting utilization are still at the initial stage, so that, more explorations are highly desired to optimize the structure and performance of TENG toward practical applications.

Integration of TENG with emerging 3DP brings new vitality and more possibilities for new energy harvesting technology. Fig. 4 summarily demonstrates several typical structure design and application of 3DP-based TENGs, including some of the rigid and flexible structure units. 3DP-based TENGs are highly desirable for next-generation distributed energy harvesters and self-powered sensing system that are expected to be integration, reliability, maintain satisfactory performance and no additional power supply. Herein, based on working modes and 3DP methods, the structural characteristics of 3DP-based TENGs are divided into two main categories, i.e. rigid and flexible structures. As mentioned above of typical structure, with the merits of simple structure, easy to fabricate, good output performance, strong practicality and extensively application. The rigid structured 3DP-based TENGs have been widely designed and reported based on FDM technology, as exhibited in Fig. 4a, b, d, e, f, g, i and j. There is evidence that these rigid structured 3DP parts are mainly used for supporting frame of 3DP-based TENGs, but uses it for functional triboelectric layer are fewer [48,49,50,51,37,40,38,53]. As shown in Fig. 4h, k, l, m, p and r, it's systematically described that the layer-to-layer structured 3DP-based TENGs were developed by DIW technology. As can be seen in the figure, flexible layer-to-layer structured 3DP parts were designed and printed by DIW methods with the liquid phase triboelectric ink materials [52,34,54,55,41,36]. In the practical application process, flexible layer-to-layer structured 3DP parts could be both triboelectric layer and as a flexible substrate layer, and have advantages in the wearable devices and implantable electronics. Composite structured 3DP-based TENGs were developed by SLS and other 3DP technologies, as shown in Fig. 4c, n, o, t and u. The composite structured parts are normally fabricated using a variety of 3DP techniques and can also be used as functional triboelectric layer for TENGs [30,56,32,42,58]. Furthermore, the flexible structured 3DP-based TENGs have been fabricated based on the SLA technology with unique advantages [33,56,35,22], which remain popular in wearable, medical rehabilitation and have remarkable advantages in wearable electronic devices, as shown in Fig. 4s, q, v and w. The 3DP have recently become popular over conventional production and processing approaches such as magnetron sputtering, imprint lithography, tape casting, spin coating, etching, electrospinning and melt blown as it offers the benefits of simple process, easy machining, high efficiency and precision. Although the 3DP is of many advantages and has been applied for the manufacturing of TENG devices, some of its defects become obstacle to further applications, such as printing resolution, aspect ratio and processing temperature. The tolerances are larger with 3DP which can lead to inconsistencies between multiple prints of the same part. When producing at larger quantities, typically these variances would not pass quality control. Another disadvantage of most 3DP is its inability to produce at a fine resolution, there will be many rigid edges on the surface of the devices that require additional finishing methods, such as sanding, to make it smooth. All this above due to the particular craft process and certain materials of this technology. Therefore, for long time, only regulatory control has been implemented in TENGs production, aiming at some particular craft processes, equipments and materials, can't satisfy the flexible request in system.

Fig. 5 summarily demonstrates the structural characteristics and functionalities of 3DP parts for fabrication of 3DP-based TENGs. Through the analysis and data statistics, found that 3DP parts are divided into two main categories, i.e., rigid and flexible structures from the aspect of structural strength. One of which the percentage of rigid and flexible 3DP parts reached 72.5% and 27.5%, respectiveiy. Overall, the relative advantage of various rigid 3D-printed parts is evident, which the percentage of actual use is high, as shown in Fig. 5a. It is mainly due to 3DP technologies and 3DP materials for fabrication of rigid structural TENGs more easily than flexible structural TENGs. Based on the practical applications, the percentage of 3DP parts with different functionalities for fabrication of TENGs, as shown in Fig. 5b. The functionalities of 3DP parts are divided into three main categories, i.e., triboelectric layer, supporting frame and other, them reached 52.5%, 42.5% and 5%, respectively. In a word, researchers have given priority to fabricate triboelectric layer by 3DP and improve output performance of 3DP-based TENGs.

Four typical working modes of 3DP-based TENGs

Based on the coupling effects with triboelectrification (i.e., contact electrification) and electrostatic induction, TENGs can be categorized into four typical working modes, i.e., contact-separation mode, sliding mode, single-electrode mode and free-standing mode, which the same is true for 3DP-based TENGs. Integration of TENG technology with emerging 3DP brings new vitality and more possibilities for energy harvestors and self-powered sensors. Typical structures and practical applications of 3DP-based TENGs are schematically illustrated in Fig. 6, which presents in the form of the four working modes. According to above four working modes and corresponding 3DP methods, while each 3DP-based TENG has its own structural characteristics, electrical performance, application occasions, advantages and disadvantages.

With the advantages of simple structure and pulse output, the contact-separation mode 3DP-based TENGs have been widely reported, while depends on relative motion between two triboelectric layers perpendicular to the interface. Their simplest structure are that triboelectric layers are directly covered the surface of 3D-printed parts and film electrodes. As shown in Fig. 6a, b, c, f, g and h, the contact-separation mode TENGs were fabricated by 3D-printed framework or coating organic dielectric film on the contact surfaces of 3D-printed parts [38,40,52,34,59,60]. The obtained devices can be used to energy harvesting and self-powered sensing, which the structural characteristics are vertical movement and a large gap. It has many merits such as high output voltage, pulse output, good performance and versatile applications and so on. It can be applied to various occasions like vibration, pressing, impacting and sharking. However, the output performance will be affected by contact velocity and dielectric thickness, and is influenced by separation distance. The sliding mode 3DP-based TENGs relies on the relative displacement and the applied force in the direction parallel to the

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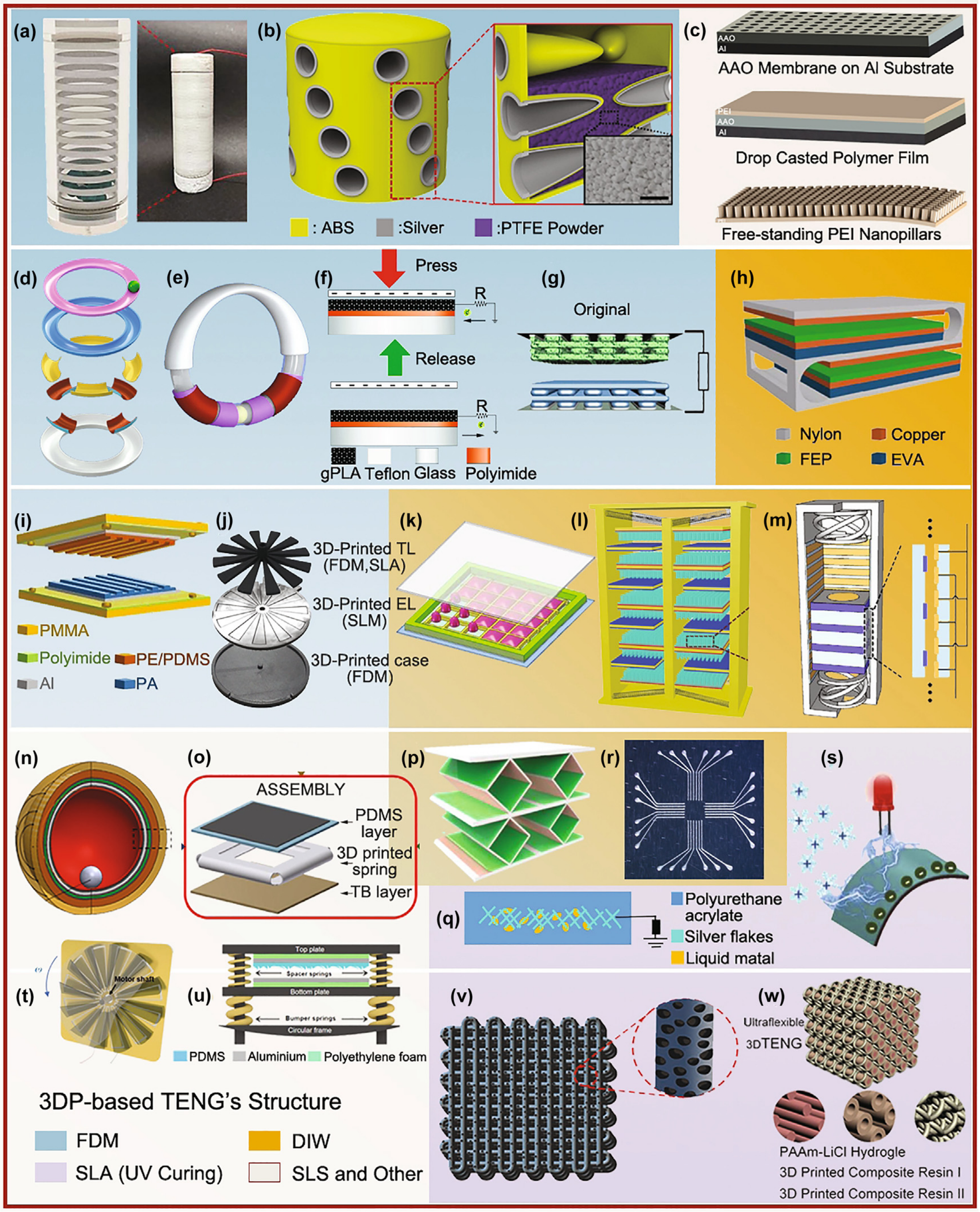

FIG. 4

Typical structure design and application demonstrations of 3DP-based TENGs. (a), (b), (d), (e), (f), (g), (i) and (j) Rigid structured 3DP-based TENGs developed by FDM technology [48,49,50,51,37,40,38,53]. (h), (k), (l), (m), (p) and (r) Layer-to-Layer structured of 3DP-based TENGs developed by DIW technology [52,34,54,55,41,36]. (c), (n), (o), (t) and (u) Composite structured 3DP-based TENGs developed by SLS and other technologies [30,56,32,42,58]. (s), (q), (v) and (w) Flexible structured 3DP-based TENGs developed by SLA technology [33,56,35,22].

interface, as shown in Fig. 6d, e, i, j and k. Their simplest structure are that two different triboelectric materials are directly covered the surface of 3D-printed parts and film electrodes, form two opposite sides of the same TENG [55,61,56,33,62]. The mode has significant structural characteristics in comparison to the contact-separation mode, such as horizontal movement, rotational movement and almost without gap, also highlighted the advantages of their own at high frequency, continuous and high electricity output. So that, can be applied to various occasions like by horizontal/rotational movement to harvest air and water flow energy. However, there are some inevitable shortcomings, such as easy damage of contact surface, poor long-term etc. In addition, single-electrode mode TENG can also through the structural optimal design for integration of 3DP-based TENGs. The simplest structure is that organic triboelectric materials are directly simply print on a electrode surface by 3D-printer. As

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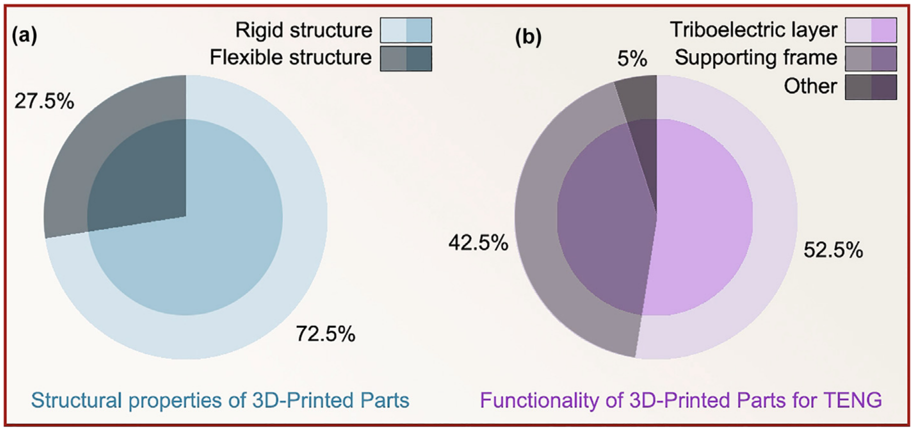

FIG. 5

Structural characteristics and functionalities of 3DP parts for fabrication of 3DP-based TENGs. (a) Structural characteristics and the percentage distribution of 3D-printed parts (into two categories: Rigid and Flexible structure) to use for fabricating 3DP-based TENGs. (b) Functionalities and the percentage of 3D-printed parts (into three categories: triboelectric layer, supporting frame and other) to use for fabricating 3DP-based TENGs.

shown in Fig. 6l, m, n, p, and s, energy harvesting unit was fabricated by printing PLA, polymeric hydrogel, PDMS, photosensitive resin or synthetic resin on the surface of conductive substrate [35,37,50,22,64]. However, the working condition of 3D-printer is relatively severe, so the property requirement for the 3DP materials is very high, the triboelectric and mechanical properties of mature materials are poor in the current application. Therefore, this related research has been a key research and attracted great attention in material field. The existing researches confirmed that single-electrode mode TENGs has the advantages of simple structure, versatile in harvesting energy, and easy for integration. Its application scenarios focus on what they are good at touching, sliding or typing. Although single-electrode mode 3DP-based TENG is easy to prepare and convenient for usage, but low and unstable output performances restrict its wide application. In order to further improve power outputs, energy conversion efficiency and working stability, single-electrode mode 3DP-based TENG is usually optimized to freestanding mode. Research indicates that freestanding mode 3DP-based TENGs can be easy to fabricated according to practical application requirements. For example, 3DP a layer of triboelectric material on the surface of interdigitated electrodes with grating structure, or attaching a layer of triboelectric material on the surface of 3D-printed frame structure and to combine a layer of grating electrodes, as shown in Fig. 6o, q, r, t and u. In addition to directly paste, functional triboelectric layer can be deposited on the surface of 3D-printed structure via various physical or chemical techniques [63,49,51,48,54]. The electrical output properties are impacted by freestanding height, velocity and electrode gap, which is suitable for multiple forms of movements suche as sliding, vibration and rotational energy harvesting. The typical electrical output characteristics of freestanding mode 3DP-based TENGs are presented that including high energy conversion efficiency and symmetric grating electrodes but charge distribution is opposite. It is found that low open-circuit voltage and high short-circuit current, especially compared to single-electrode mode 3DP-based TENGs. Furthermore, the main drawback to it is the cost, easy to wear at a high sliding speed operation. Therefore, some wear-resistant materials and optimal structure of designs are gradually used to enhance the wear-resisting property of energy harvesting devices. Overall, the working principle and structural characteristic of 3DP-based TENGs are demonstrated, it will make a big influence on areas like portable electronics and self-powered sensing system, and so on.

Basic triboelectric mechanisms of 3DP-based TENGs

Triboelectric effect is the phenomenon that a physical contact between two dielectric materials causes triboelectric charges on the two surfaces, as a powerful new technology for converting mechanical energy into electricity based on the coupling of triboelectrification effect and electrostatic induction effect [65–72]. So far, it is found that the triboelectric mechanisms of the four working modes of 3DP-based TENGs have been demonstrated through theoretical calculation and experimental data. It is conformable to the results obtained that the electron transfer instead of ion transport is the dominant process of triboelectrification [73–76]. Nevertheless, each working mode has its own structural characteristics, law of surface charge accumulation and the distribution of TENG's four working modes in field of application occasions, which is summarized and compared in Fig. 7. The the contact-separation mode 3DP-based TENGs depends on practical effects of relative motion between two triboelectric layers perpendicular to the interface like separation distance, velocity and applied force (Fig. 7a), while the sliding mode more relies on the relative sliding displacement and velocity in the direction parallel to the interface (Fig. 7b). The single-electrode mode takes the ground as the reference electrode and is versatile in harvesting energy from a freely moving object without attaching harvesting energy from a freely moving object without attachingan electric conductor (Fig. 7c). The freestanding mode consists of a triboelectric layer with grating segments and two stationary electrodes with interdigital patterns (Fig. 7d).

Anyhow, the characteristic of charge generation and transfer mechanisms with the various mode 3DP-based TENGs are similar, that they all follows the fundamental principles of triboelectric mechanisms [77,78]. Therefore, if our comprehensive

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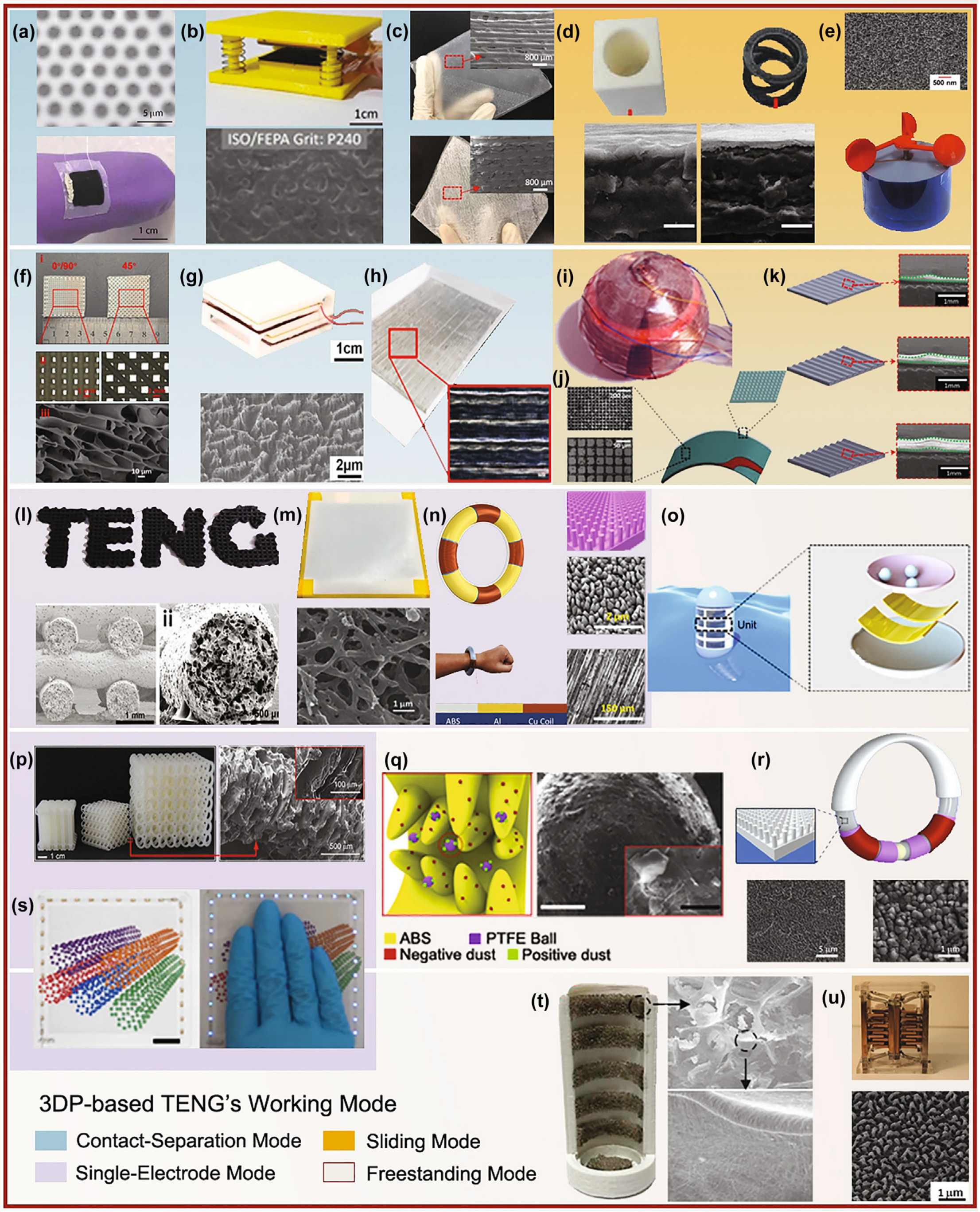

FIG. 6

Typical structure design and application demonstrations of 3DP-based TENGs based on four working modes. (a), (b), (c), (f), (g) and (h) Typical structure design and application demonstrations of contact-separation mode 3DP-based TENGs [59,60,38,40,52,34]. (d), (e), (i), (k) and (j) Typical structure design and application demonstrations of sliding mode 3DP-based TENGs [55,61,56,33,62]. (l), (m), (n), (p), and (s) Typical structure design and application demonstrations of single-electrode mode 3DP-based TENGs [35,37,50,22,64]. (o), (q), (r), (t) and (u) Typical structure design and application demonstrations of freestanding mode 3DP-based TENGs [63,49,51,48,54].

analysis on any one of these modes, and suffices to illustrate the point. For instance, the charge generation and transfer process of a typical the contact-separation mode 3DP-based TENG is investigated (Fig. 7a). By basic triboelectric mechanisms, originally, when a physical contact between two triboelectric materials because of mechanical motion, that causes positive and negative triboelectric charges on the two surfaces [75]. At this juncture, the equal numbers of positive and negative charges are induced on the bottom and top electrodes, respectively, due to the electrostatic induction effect [74]. Secondly, a relative separation between the two as caused by sustained mechanical motion results in an electric potential drop across the two electrodes built below the triboelectric materials. When they are separating and gradually moving away, which drives the electrons to flow

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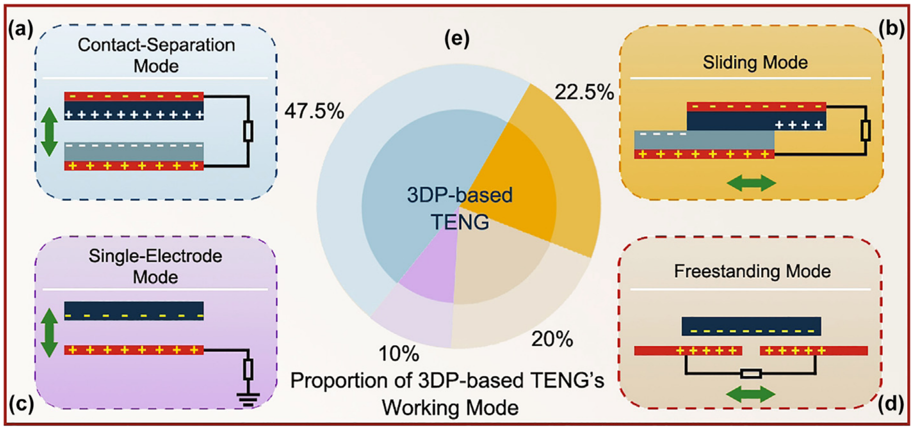

FIG. 7

Basic triboelectric mechanisms of 3DP-based TENGs with four working modes, and proportion of TENG's four working modes in field of research. The red model refer to conductive electrode materials, while the smalt, gray models represent triboelectric materials, which both materials fabrication can be made by 3DP.

between the two in order to balance the electrostatic system. In completely separated cases, the positive and negative triboelectric charges are fully equilibrated, reflecting the conservation of charges in this period. It is noteworthy that the accumulated charges will not be entirely annihilated due to the innate features of dielectric triboelectric materials, which they will be maintained for a sufficiently long time. And then, when a reverse motion, the two are approaching back to each other, the accumulated induced charges will flow back through the external load to compensate for electrical potential differences. Finally, after the whole system returns to the initial state, the positive and negative triboelectric charges are fully offset again. As a result, a contact-separation process between the upper and lower triboelectric materials will generate an instantaneous alternating potential and reciprocating current through the external load.

Meanwhile, one important point is that the polarities of both two triboelectric materials depend on their ability to gain and lose charges, which the process of gain or loss can also be described on the atomic or molecular scale by electron cloud-potential well model [74,79,80]. Prior to contact between the two triboelectric materials, their respective electron clouds remain separated and without overlap. In this case, the electrons are firmly bound and prevented them from freely escaping, this due to electron cloud-potential wells have not been broken. When the two contact, the electron clouds collide with each other to form ionic or covalent bonds that the initial balanced potential well is broken [81–85]. Therefore, there occur charge transfer between the two triboelectric materials which leading to electrons hopping from one with higher energy to its counterpart [86–89]. After once again separated, the transferred electrons remain but the thermionic emission of triboelectric charges occurs, resulting in partial release of the electrons from one of the triboelectric materials, while simultaneously the other one triboelectric material will be replenished from the environment electrons [90–95]. In terms of the morphology (into two categories: solid and liquid), structural features (into two categories: rigid and flexible) and functionalization (into three categories: triboelectric layer, supporting frame and other) of triboelectric materials, various approaches have been undertaken. The most common polymers considered by researchers include PLA, TPU, PDMS, PUA, PTFE, ABS, PET, PCL, and so on. The tensile strength, flexibility, durability and triboelectric performance of these materials determine them suitable for which kind of the operating modes and principles of TENG applications.

Electrical output performances of 3DP-based TENGs

Based on the four modes illustrated above, we have statistical analysed the performances of various 3DP-based TENGs depending on specific applications. The electrical output performances are also summarized and compared, as shown in Fig. 8. The rangs of performance mainly covers open-circuit voltage ($\boldsymbol{V}_{oc}$, V), short-circuit current ($\boldsymbol{I}_{sc}$, μA), the amount of charge transferred ($\Delta\boldsymbol{Q}_{sc}$, nC), instantaneous power ($\boldsymbol{P}$, mW), the surface charge density (ρ, μC/m^2), and the density of peak pulse power ($\boldsymbol{P}_D$, W/m^2). To date results showed that the output performance of 3DP-based TENGs have reached a higher level [30,37,38,41,56,57], such as the rangs of maximum values were 1900 V, 6140 μA, 2580 nC, 510 mW, 120 μC/m^2, and 104.6 W/m^2, respectively. Generally speaking, the contact-separation mode 3DP-based TENGs have higher electrical outputs and better practicability than those with other mode [96,97]. For 3DP-based TENGs, the electrical output performances are one of important specifications for evaluating quality and latent commercial value undoubtedly. Firstly, enhancing the triboelectric properties of 3DP material is one of the important strategies to improve the electrical output performances. Secondly, optimizing the structures of 3DP-based TENGs to enhance the conversion efficiency that is a key technical means for this. And finally, promoting the manufacturing level of the devices and further refining the energy management circuit, as it has to be.

Furthermore, the published works have demonstrated that the developed 3DP-based TENG devices have a relative stability, that is the foundation for its practical applications and a major challenge. In which will be influenced primarily by 3DP

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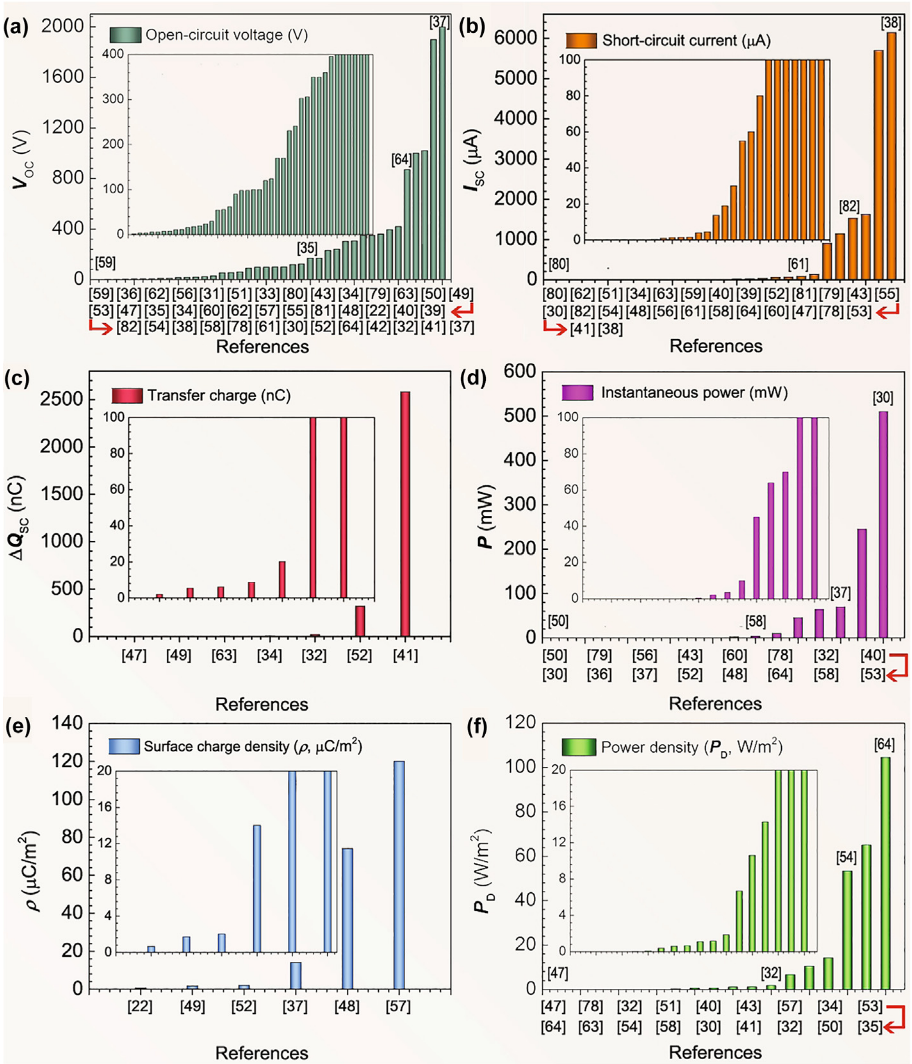

FIG. 8

Summary and comparison of the electrical output of various 3DP-based TENGs with four working modes, mainly including V_{oc}, I_{sc}, ΔQ_{sc}, P, ρ, and P_D.

technological parameters and its input materials. This is because all these factors (such as material selection, structure design, printing resolutions, deposition rates, printing axis orientation, printing layout and defaults layer thickness, printing temperature setting, etc.) are affecting the stability in printed part. Further, internal stress and deformation can occur during 3DP process, which these factors have an undesirable influence on the dimensional accuracy and mechanical properties of 3DP-based TENG devices. To augment the stability of 3DP-based TENGs, at first, these factors have to be carefully controlled during 3DP process. Despite the stability of 3DP-based TENGs demonstrated so far offers impressive applicability to drive portable electronics automatically operation, but still is one of the key technical challenges of this technology in practical application. Fortunately, that is exactly what it also provided a much broader researching space for researchers.

Although, the electrical output performances of the current 3DP-based TENGs with four working mode are summarized and compared, which the result did not reach statistical significance. Due to different triboelectric materials, device structures and sizes, varied loading conditions and environmental factors, it is rather difficult or even impossible thorough to distinguish good or bad between different 3DP-based TENGs. However, the result promising that deserves prompt attention. For various structures of 3DP-based TENGs and its corresponding output characteristics, some representative application instances in terms of these ranges are presented in Fig. 9. It has been proved that 3DP-based TENGs can be applied in almost every field involved power sources and sensor networks, as well as has largely enriched the ranging of energy harvesting from human activities and natural environment, etc. In order to provide more qualitative and quantitative information for future research and practical applications, they are still worth introducing here. "For example, self-powered sensing based on various human kinetic characteristics which the demand of voltage and power is below the average 10 V and 0.01 ~ 0.3 mW, such as light, pressure, sensor, and acceleration sensors have been adopted in many fields. Furthermore, 3DP-based TENGs have been regarded as effective tool to harvest mechanical energy that is ubiquitous but usually wasted in everyday life, which

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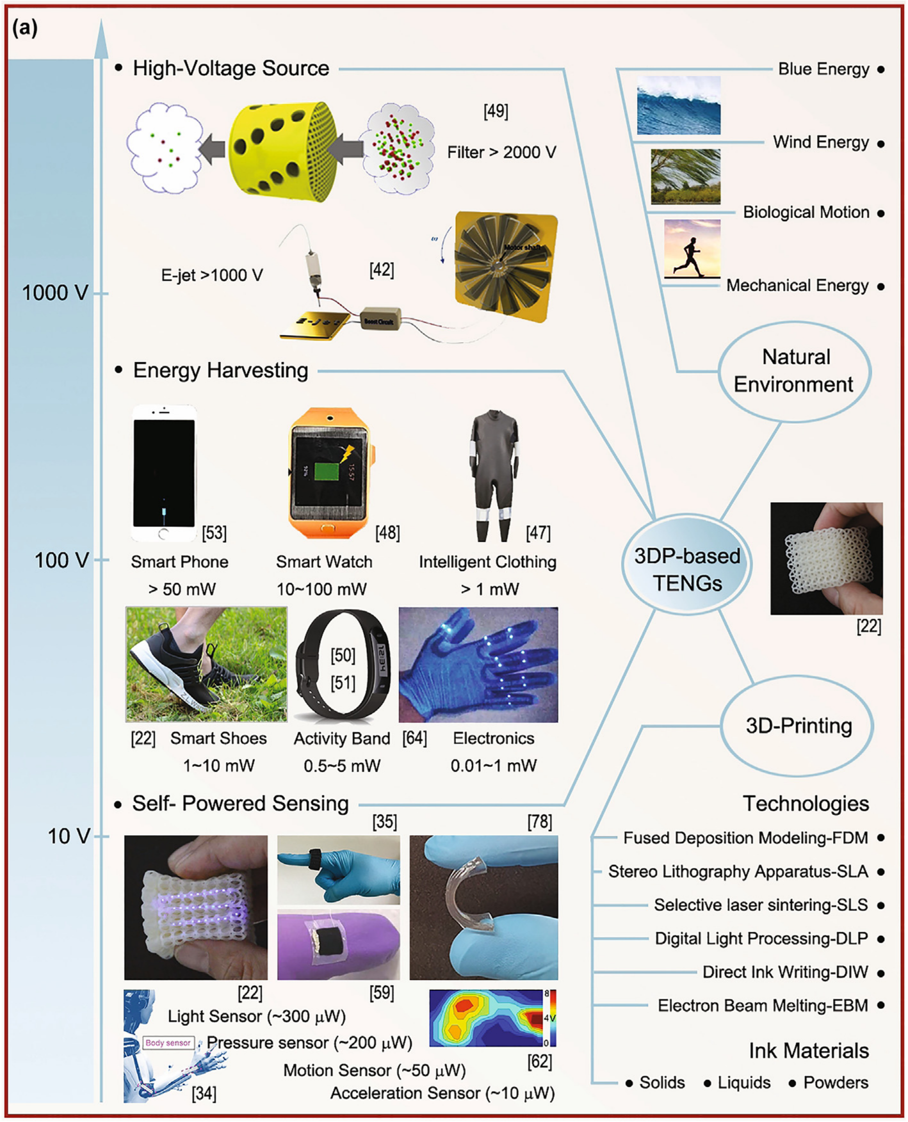

FIG. 9

Representative application instances of 3DP-based TENGs and its corresponding electrical output characteristics.

have broad application prospects in wearable power supply, such as smart shoes, band, phone, watch, intelligent clothing, and low-power electronic devices (probably somewhere the rang of less than 100 V and 0.1 ~ 50 mW). Therefore, it is possible for us to realize the energy utilization in a self-sufficient way, to achieving the seamless combination of 3DP-based TENGs and human motions. In term of high voltage source (for instance more than 1000 V), several kinds of emerging applications have already been developed, such as air purification, electrospinning, intelligent prosthesis, and triboelectric soft robotic. Although some satisfactory results have been achieved about the applications of 3DP-based TENGs, it is still in its infancy and many difficulties need to be overcome. However, there is no doubt that the application of 3DP-based TENGs in all fields is an inevitable trend. In future, it can be forecasted that 3DP-based TENGs will become an important part of energy harvesting from nature environment."

Discussion and summary

In summary, the 3DP-based TENG is the fusion of modern additive manufacturing and TENG technology. As a newly developed energy technology that 3DP-based TENGs have shown strong vitality and enormous advantage in various fields of future society, which greatly promotes the developments of energy harvesting and self-powered system. In order to make an overall grasp with its current situation, provide with reference for the future research and development of TENGs by using 3DP technologis. It is necessary to make a general introduction and discussion about what the basic knowledge, current research progresses and future development trends of 3DP-based TENGs as well as their new harvesting forms for various types of environmental energy here. This paper reviews the updated progress in studying 3DP-based TENGs on mechanical energy harvesting and multi-functional self-powered sensing. A brief overview of the combination of the two from the 3DP technologis and TENGs is

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conducted, a comprehensive investigation on output performances and practical applications of current 3DP-based TENGs have been overviewed and its advantages and disadvantages are also discussed in detail.

It is well known that the ubiquitous triboelectric effect endows TENGs with more diversified structure and wide material choices, as well as more significant that the design possibilities based on 3DP are protean and endless. The energy and sensor technologies based on 3DP-based TENGs will significantly impact the development of internet of things, portable electronics, robotics, and artificial intelligence and human–computer interaction, from which we can see the future prospects of 3DP-based TENGs. Herein, some promising and representative application occasions in terms of these fields are presented in Fig. 10, which will impact the world for the future, the unique advantages are demonstrated as follows:

(1) *Unique device structure:* Given the vast potential and applicability of 3DP-based TENGs, one is that owes a good deal to their unique structure. Such as single (1D), thin film (2D) and bulk stucture (3D) on the spatial dimensions, as well as like grid shaped and multi-layer, also like flexible, stretchable, elastic and rigid, etc. And it all has benefited from 3DP offers accurate architectures ability for the various customized TENG units, in order to cope with different requirements of application occasions.
(2) *Infinite design space:* 3DP is a process of making three dimensional solid objects from a digital files, which provide a large-area integrated new strategy and comprehensive service platform for the structure design and optimization of TENGs. The basic TENG structure units can be adjusted based on the feature of mechanical energy in nature, which make its performance better and its

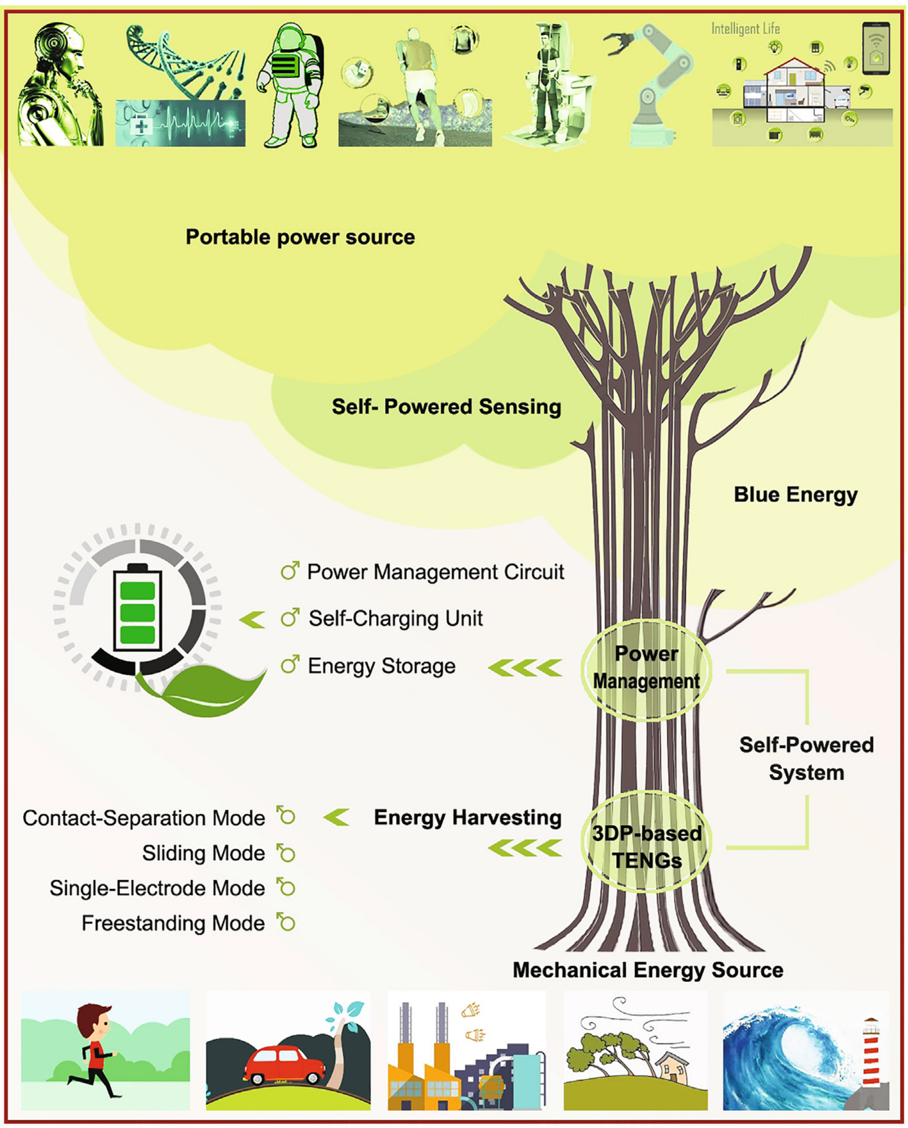

FIG. 10

Promising application fields of 3DP-based TENGs in portable power source, self-powered sensing and blue energy, in which self-powered system that harvest energy from natural environment, managing the input power, and effectively store the harvested energy for sustainable driving of distributed electronics.

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structure more proper, and help 3DP-based TENG device is brought closer to the real-life user. As well as the further researches and more popular applications of TENGs are full of infinite possibilities due to 3D digital design, computer-aided analysis and process function, dummy manufacture etc.

(3) *Implementable integration process:* The main advantages of 3DP for the fabrication of TENG is reflected not only in compatibility, diversity, high efficiency, large area and mass customization, but also more importantly increases accuracy, stability and reliability of 3DP-based TENGs. In additon, it enables reducing material usage waste, minimizing human intervention as well as provides highly automated and safety production facilities. That could make the 3DP-based TENG products be more closer to practical applications and arouse the consumers's resonance, and consequently realize the value of portable and distributed energy better.

Although great achievements related to 3DP-based TENGs have been achieved in terms of research and all kinds of applications for energy harvesting and self-powered sensing. It is also anticipated that this technology will have a significant impact that will provide a realistic solution of self-powered energy system for commercial electronics. Yet the greater disparity still exists between the experimental development and practical commercial applications. Herein, potential difficulties and obstacles for the widespread commercial applications of 3DP-based TENGs are also analyzed and discussed from 3DP technologies and materials, electrical output performances and mechanical properties. Such as the perspectives of energy conversion efficiency, output power, stability, durability, wearability and target market. As well as the current one of the important challenges for 3DP-based TENGs is how to integrate electrode layer and functional triboelectric materials, for example, by optimizing the 3DP technologies and materials, integration process. Striving continue to towards the progress of efficient, reliable and practical 3DP-based TENGs and their prospects, the following aspects are indispensable and the still greater effort should be provided to overcoming these challenges.

(1) *3DP technologies:* To date only the FDM technologies have been utilized for fabricating 3DP-based TENGs frequently, while but the use of other approaches are fewer such as SLA, DIW and SLS. Meanwhile, current most 3DP technologies can be used to integrate TENG devices but not quite matching, while are incompatible with relative mature stucture of TENG and its fabricating process. It should be noted that the currently used 3DP are relatively single manner with the lack of integrated and multifunctional multi-process technique as primarily methods. So that, the future work should try to combines the use of other approaches, such as digital light processing (DLP), electron beam melting (EBM) flexible planographic and multimaterial multinozzle 3DP, etc. All of these are crucially important for solving specific structure and complex applications.

(2) *3DP materials:* The current 3DP materials can print out something rigid, flexible, stretchable and biocompatible structures for TENGs. Unfortunately, some 3DP-based TENG devices failed to meet requirements of practical application due to costs and mechanical properties. Such as the flexibility, stretchability, wearability and mechanical strength of materials are unable to cope with complex working conditions, extreme deformations and continual loadings. Moreover, the shortcoming is its expensive price, and the material system is still not perfect that not every triboelectric or conductive material are available on 3DP platform. Therefore, future research should commit to the development and optimization design of 3DP material systems for overcoming these challenges. We should also strive to fabricate 3DP-based TENGs by 3DP platform, to enable more suitable for various working conditions, more extensive scope of application.

(3) *Electrical output performances:* At present, despite 3DP-based TENGs with a certain level of triboelectric property, but output powers are far less than the actual demand of most micro-rnano electronic devices, so make them difficult to applied in practical production and daily life. Fortunately, it is demonstrated that there are several approaches to improve the energy conversion efficiency of 3DP-based TENGs, such as increasing effective contact area, optimizing device structure and power management circuits, improving triboelectric property of 3DP materials, etc. In addition considering the application environment, the effective encapsulation is essential for stable and efficient of electrical output performances. Therefore, it is firmly believed that the output powers of future TENGs will be greatly enhanced, even better than the power requirement of traditional electron devices.

(4) *Target market and prospects:* Even if it can be well resolved and coped with the above challenges which ensure the high quality for them, yet the wide commercial applications of 3DP-based TENGs are still influenced by a lot of the internal and external factors. In the future market of portable power supplying and self-powered sensing system, whether 3DP-based TENGs can stand out among various solutions, not only still depends on their quality, cost and core technologies, but also market demand, competition, customer's value identity and orientation and marketing. In order to improve the future competitiveness of 3DP-based TENGs, the researchers still must always focus on enhancing its comprehensive performance and reducing its cost.

In summary, while challenges remain, and what's more 3DP-based TENG has a bright future, which their vigorous development and widespread application are an irresistible trend of a high-tech new era. It is firmly believed that the current critical challenges of 3DP-based TENGs will be well overcomed with the progress of science and technology. In a word, it is expected that 3DP-based TENGs will continue rapid development in the coming a decade or two, as well as some predictable applications will commence to play a vital role in traditional markets, possibly

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creating a dominated industrial chain based on 3DP-based TENGs.

Declaration of Competing Interest

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

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