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Self-powered flexible electronics beyond thermal limits

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ABSTRACT

Self-powered flexible electronics using high-performance inorganic materials have been studied and developed for the essence of future electronics due to the thing, lightweight, self-sustainable, and biocompatible characteristics, which can be applied to body sensor network and next generation Internet of Things (IoT). However, most of inorganic materials should be processed in the high-temperature processes such as the semiconductor fabrication, which is not compatible flexible plastic substrates. Therefore, the new approaches must be demonstrated to overcome the thermal limits of previous methodology and achieve the flexible inorganic electronics on various flexible plastic substrates. In this review paper, we introduce the recent progress of technologies to realize flexible and high-performance inorganic electronics on plastic substrates over the thermal limits, i.e., laser-assisted procedure, chemical or mechanical exfoliation approaches. They are compatible not only to flexible plastic substrates but also to conventional device processes. We also explain the novel application devices such as flexible optoelectronics, flexible large-scale integration (LSI) devices, flexible energy harvesters, and flexible sensors using the recent-developed technologies beyond the previous thermal limit. This paper highlights the proper direction to complete future flexible inorganic electronics for high-performance self-powered systems.

Self-powered flexible electronic systems have been spotlighted as a powerful solution to realize system-on-plastic (SoP) without bulky energy sources, enabling sustainable, maintenance-free and independent operation of integrated flexible device components [1–15]. They have significant potential shifting the paradigm of conventional bulk silicon technology to innovative electronic fields from internet of things (IoT) to wearable sensors by resolving power consumption issues and providing human-friendly interfaces (e.g., portability, light weight, conformal contact capability)[4,16–22]. For instance, the self-powered wireless sensor network (WSN) SoP can be implanted in curved and dynamic surfaces of the organs to monitor the biological vital signs based on real-time/bilateral data transmissions without requiring infectious surgery of the battery replacement of a million sensor nodes [14,19,20,23–25].

A number of research groups have attempted to utilize organic and carbon materials via low-temperature fabrication to demonstrate self-powered flexible electronics [26–29]. However, their insufficient material properties, low durability and non-compatibility with complementary metal-oxide-semiconductor (CMOS) processes have restricted the range of applications for high-density SoP with excellent performance [30–35]. Although inorganic materials can be employed for fully functional electronic devices by using high-temperature thermal processes (e.g., semiconductor fabrication), flexible plastic substrates have inherent thermal limitations such as low melting point and heat shrinkage/expansion, which demands new concepts for implementing high-performance inorganic-based self-powered SoP [36–38].

Laser technologies (e.g., low-temperature polycrystalline silicon (LTPS), light-material interaction) have attracted considerable attentions as a novel strategy for realizing flexible electronic systems via its exceptional capability to excite rapid and non-equilibrium thermal reactions within localized space without damage of polymers [39–45]. Jeon et al. [46] reported the laser crystallization of hybrid perovskite for solar cells on a plastic substrate using a near-infrared (NIR,

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wavelength = 1064 nm) laser. In addition, inorganic-based laser lift-off (ILLO) has successfully demonstrated various flexible devices such as thin film transistors (TFTs), sensors, light-emitting diodes (LEDs) and memory devices on plastics [47–50]. From the perspectives of a broad spectrum and low-cost light source, Park et al. [51,52] utilized a flash lamp to develop high-performance flexible conducting materials and electronics.

The transfer technology including physical exfoliation and chemical etching is another promising technique to overcome the thermal limits of plastics for self-powered SoP [53–59]. Through the transfer methodology, electronic systems that are initially fabricated on a rigid substrate can be transferred to flexible plastic substrates by specifically detaching ultrathin upper layer of the rigid substrate with entire devices, and then subsequently placing them onto the plastic substrate [60–63]. This process can use conventional high-temperature annealing, nanofabrication and multi-alignment steps, whereas it is indeed possible to make them flexible on plastics. Therefore, the significant advantages can be taken for commercial SoP applications such as flexible optoelectronics, flexible large-scale integration (f-LSI) circuit and energy device [64–66].

Here, we introduce brief overview of representative technological advancements for self-powered flexible electronic systems beyond its thermal limit. In particular, we focus on the approach and mechanism of laser or transfer technologies that have played essential role in resolving high temperature issues for implementing high-performance SoP, as presented in Fig. 1. This review has four main categories: i) the laser technology including light-material interaction and ILLO, and their key applications, ii) flexible optoelectronics for biomedical devices, iii) electronics for f-LSI, iv) self-powered flexible energy harvesters and sensors. Finally, we clearly summarized the main technologies and mechanisms of the studies as a table. These new perspectives will provide a promising outlook and strategy to overcome the challenging obstacles about thermal limits in realizing the various self-powered SoP ranging from wearable sensor to healthcare system.

1. Laser technology

1.1. Light-material interaction

Light-induced photothermal interaction that converts photonic energy to thermal energy has been extensively employed for the ultrafast and selective heating of metal, inorganic and hybrid materials without damage of neighboring structures [67–70]. It has contributed to a tremendous breakthrough in opening an era of materials and electronics for self-powered SoP such as flexible energy devices, conductors and optoelectronics [67–70]. Left schematic image of Fig. 2a illustrates the laser crystallization of $CH_3NH_3PBI_3$ ($MAPBI_3$) for perovskite solar cells. [46] A Nd: YAG laser (1064 nm wavelength) was irradiated to the precursor film deposited onto indium tin oxide (ITO)-coated poly-(ethylene-2,6-naphthalate) (PEN) substrates. The laser-induced heating generated at interfacial ITO layer converted the $MAPBI_3$ into light-absorbing crystalline perovskite structure without thermal damage of polymer substrate. As shown in right panel of Fig. 2a, inverted-type flexible solar cells with 8.0% efficiency was demonstrated through optimized laser scanning conditions, resulting from the uniform and large grain-sized perovskite film.

Choi et al. reported the phase separation of silicon carbide (SiC) solid-state binary compounds induced by pulsed excimer (XeCl, wavelength of 308 nm) laser [71]. By the non-equilibrium and instantaneous (nanosecond-scale) light-material interaction, the graphitization mechanism was observed, which had been challenging due to the ultrafast phase separation and the complexity of the compound materials. Fig. 2b presents the cross-sectional high-resolution transmission electron microscopy (HRTEM) image of a 4H-SiC surface exposed by a single pulsed excimer laser, showing distinct four layers composed of randomly stacked carbon, Si, 3C-SiC and 4H-SiC, sequentially. As described in Fig. 2c, the laser-induced melt-mediated phase separation phenomena of SiC was confirmed by molecular dynamics (MD) simulations. The studies on SiC phase separation provide an important technology and concept that will be extensively utilized for the synthesis of ultrathin nanomaterials on plastic via the light-induced decomposition of a binary system.

Xenon flash lamp has received a great deal of attentions because of its high light-output efficiency, rapid and large-area processability as well as roll-to-roll production compatibility [41,72–74]. Park et al. demonstrated flash-induced plasmonic interactions of Ag nanowires (NWs) for high-performance flexible transparent conducting electrodes (FTCEs) with strong adhesion on a plastic substrate [51]. As illustrated in Fig. 2d, the ultraviolet (UV) and near-infrared (NIR) light emitted from broad spectrum simultaneously induced intensive heat at the Ag nano-junctions and along the bottom NWs, respectively. TEM image of the Fig. 2e presents a fully welded Ag NW junction by extreme hot spot generated from UV light of the flash lamp, which enables the FTCEs with exceptionally low sheet resistance of $5\,\Omega\,sq^{-1}$ at high transparency of 90%. Scanning electron microscope (SEM) image in the Fig. 2f shows the locally melted interface between Ag NWs and polyethylene terephthalate (PET) polymer, enhancing the adhesion force of the NW network by 310% compared to that of pristine Ag NWs.

The flash-induced welding technique was further advanced via the flash spectrum tuning concept, which was based on plasmonic properties of nanomaterials [52]. Park et al. initially performed the polarization and spectrum dependent simulations to calculate the heat generation near the Cu NW junction, as presented in Fig. 2g. A strong field enhancement at the NW junction occurred when the incident electric field with 600 nm wavelength was perpendicularly polarized to the top Cu NW, indicating that intensive heat could be produced at the nano-

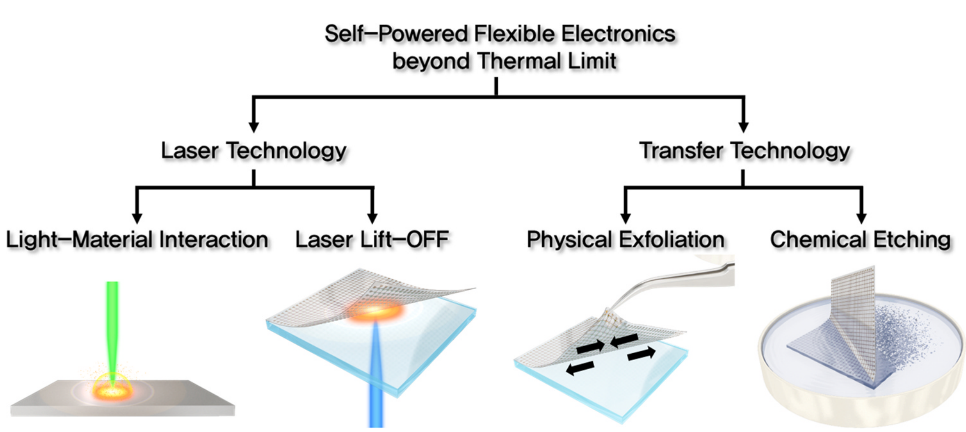

Fig. 1. Illustration of representative strategies for realizing self-powered flexible electronic system beyond the thermal limit.

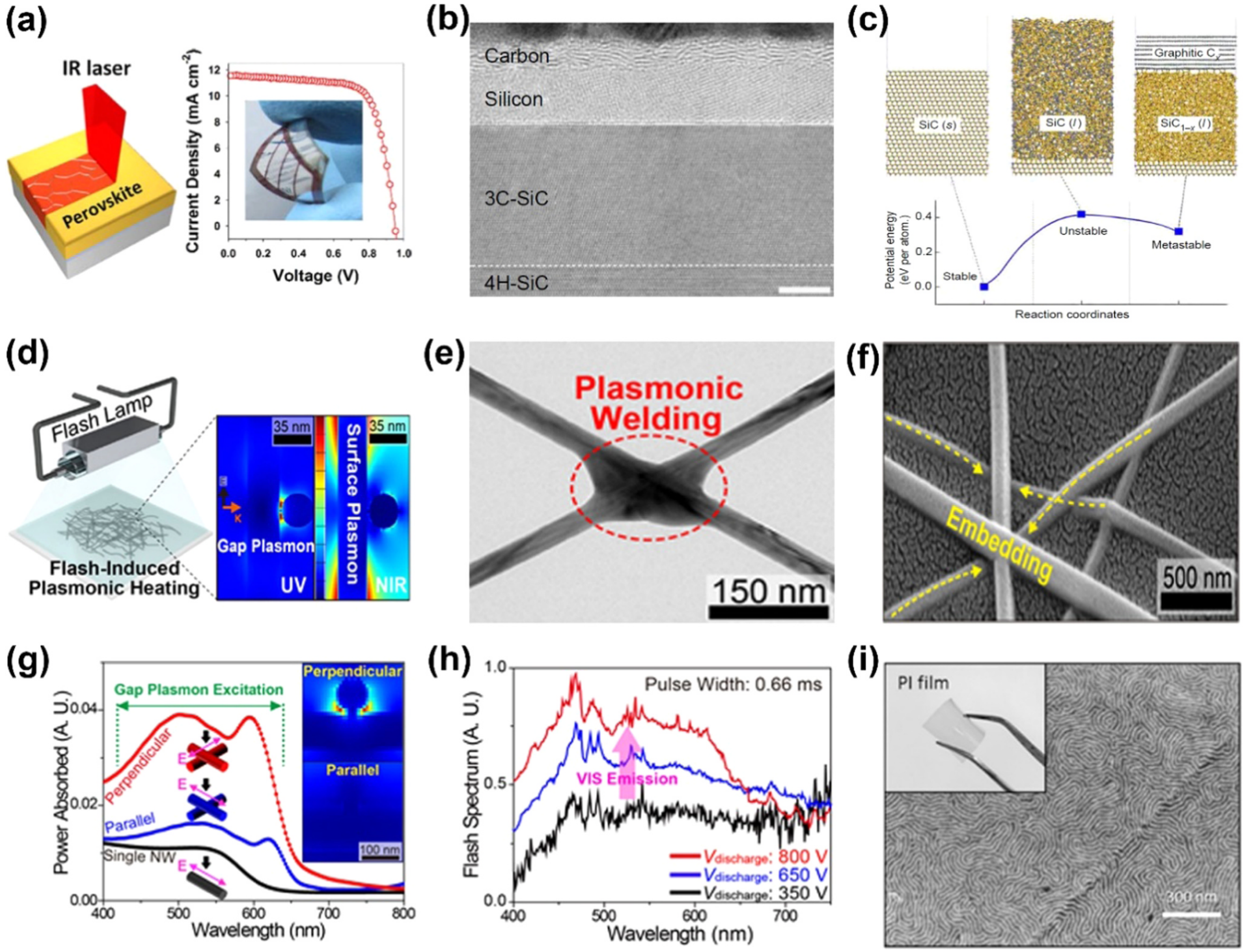

Fig. 2. (a) Schematic description of $MAPBI_3$ laser crystallization for flexible perovskite solar cells (left). J – V curve of a solar cell fabricated on ITO deposited PEN substrate (right). Inset shows photographic image of the flexible solar cell demonstrated by the laser crystallization technology. Reproduced with permission from [46]. Copyright 2016, American Chemical Society. (b) HRTEM image of 4H-SiC surface after single-pulse irradiation. Scale bar, 5 nm. (c) MD simulations for observing phase separation phenomena of 4H-SiC. Reproduced with permission from -SiC. [71]. Copyright 2016, Springer Nature. (d) Schematic of the Ag NW flash lamp processing, and local field distribution generated near the Ag NW junction under UV and NIR light. (e) A TEM image of a fully welded Ag NW junction. (f) SEM image of the embedded Ag NWs into the PET film. Reproduced with permission from [51]. Copyright 2017, John Wiley and Sons. (g) Cu NW finite-difference time-domain simulations from 400 to 500 nm light wavelength region. The inset presents the field distribution near the Cu NW junction generated by the 600 nm wavelength flash light polarized perpendicular or parallel to the first NW. (h) Normalized spectrum of flash light irradiated by various discharging conditions (350, 650, and 800 V at a 0.66 ms pulse duration). Reproduced with permission from [52]. Copyright 2017, John Wiley and Sons. (i) Self-assembled BCP on a CMG/PI substrate by flash light annealing. The inset shows the photo of the BCP on a flexible CMG/PI after flash irradiation process. Reproduced with permission from [75]. Copyright 2017, John Wiley and Sons.

intersection for favorable NW welding. To maximize the plasmonic welding of Cu nano-junctions, the spectrum of flash light was tuned based on the photothermal properties of Cu NWs by adjusting the discharging voltage and pulse duration of the flash lamp ignition. As shown in Fig. 2h, the high-intensity visible light could be irradiated to Cu NWs through the optimal flash emission condition (discharging voltage: 800 V, and pulse width: 660 µs), which facilitated a low-cost and high-performance Cu NW network without any oxidation. Jin *et al.* demonstrated ultrafast self-assembly of block copolymers (BCP) with a high Flory-Huggins interaction parameter (χ) on a flexible substrate by pulsed flash light irradiation.[75] Fig. 2i presents the morphology of self-assembled high-χ BCP on the chemically modified graphene (CMG)/polyimide (PI) film, showing sub-10 nm scale nanopatterns. Photothermal conversion generated at CMG layer induced instantaneous BCP heating/quenching, and then sequentially the BCP assembly was well-established without damage of the bottom polymer due to the millisecond-timescale of flash light irradiation.

1.2. Inorganic-based laser lift-off

ILLO has been widely applied to transfer entire devices onto flexible substrates to implement numerous electronics including energy and computing devices for self-powered SoP [47,76–78]. When a laser is irradiated from the backside of the rigid transparent substrate, light can pass through the bulk substrate and reach to the laser absorbing sacrificial layer, causing various photothermal reactions such as dissociating, explosive gas releasing, and melting/vaporizing. Consequently, the interfacial adhesion between an active electronics and the substrate is weakened, which enables ultrathin device layer to be safely delaminated from the rigid substrate without any cracks, wrinkles and mechanical deformations [36,79]. This ILLO process has considerable merits for demonstrating multifunctional and high-performance flexible electronics without problematic issues of nanoscale alignment and fabrication on plastics since it can adopt conventional high-temperature CMOS processes.

Kim et al. implemented flexible thermoelectric generators (f-TEGs)

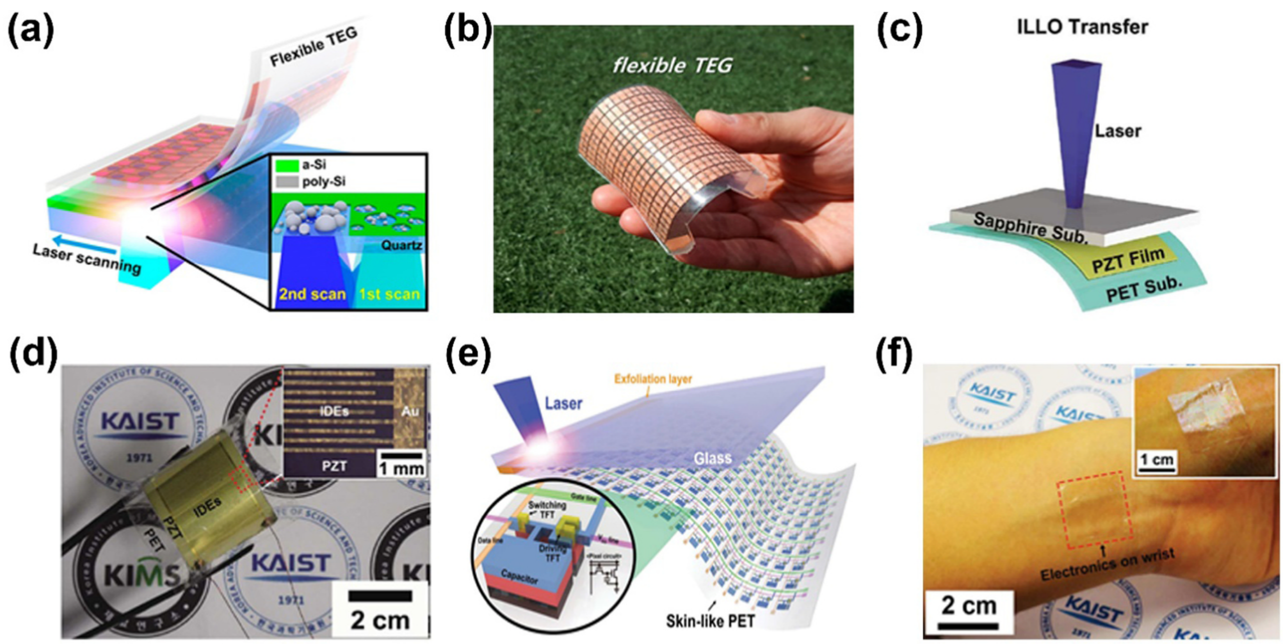

Fig. 3. (a) Schematic illustrations of the LMS-based ILLO method for the f-TEG. (b) Optical image of the freestanding f-TEG demonstrated via a LMS lift-off. Reproduced with permission from [80]. Copyright 2016, American Chemical Society. (c) Drawing image of the flexible AD PZT energy harvester enabled by ILLO fabrication process for self-powered WSN. (d) Photograph of the PZT harvester on PET substrate bent by tweezers. The inset presents a top-view microscopic image of the PZT energy harvester integrated with IDEs. Reproduced with permission from [20]. Copyright 2016, John Wiley and Sons. (e) Schematics of an ultrathin oxide TFT array demonstrated via the ILLO for transparent display, consisting of a capacitor, a driving TFT, and a switching TFT. (f) Photographic image of the skin-like TFT device array conformally attached to a human wrist. Reproduced with permission from [48]. Copyright 2016, John Wiley and Sons.

as a power source via the laser multi-scanning (LMS) lift-off process, as schematically illustrated in Fig. 3a [80]. After fabricating an original TEG on a rigid SiO_2/a-Si/quartz substrate, a pulsed XeCl excimer laser was irradiated several times to the sacrificial dehydrated amorphous Si (a-Si) layer through the backside of transparent quartz substrate, successfully reducing the adhesion between TEG device and the mother quartz substrate by inducing the agglomeration of the a-Si sacrificial layer. The exfoliated f-TEG module in Fig. 3b exhibits outstanding output power density ($4.78\,mW\,cm^{-2}$ at $\Delta T = 25\,°C$) as well as high mechanical reliability without degradation of the device performance.

Fig. 3c shows a schematic of the ILLO transfer step for flexible piezoelectric ceramic thin film [20]. The aerosol-deposited Pb $(Zr_x,Ti_{1-x})O_3$ (AD PZT) was transferred from a sapphire substrate onto a plastic substrate by the laser-induced local vaporization of the interface between the sapphire substrate and a PZT ceramic film. The demonstrated flexible piezoelectric energy harvester via the ILLO process generated an excellent current level of 35 µA and a voltage output of 200 V (Fig. 3d).

Lee et al. implemented high-performance and transparent oxide TFT array on an ultrathin (4 µm thickness) PET film via the ILLO process, as schematically illustrated in Fig. 3e [48]. Upon exposure of the XeCl laser to the a-Si:H layer, the optically reactive film subsequently released hydrogen gas with massive volume, which reduced the adhesive force between the a-Si:H sacrificial layer and the glass substrate without thermal and structural damage of the device array. As described in Fig. 4f, the skin-like indium zinc oxide-based TFTs could be conformally attached onto a human wrist, with the high effective mobility of $\sim 40\,cm^2\,V^{-1}\,s^{-1}$ and mechanical stability.

2. Flexible optoelectronics

2.1. Inorganic-based micro light-emitting diodes

Inorganic-based microLEDs have recently attracted significant attentions for the self-powered flexible displays and biomedical applications due to their outstanding optical output, excellent power efficiency, and long lifetime [5,81–87]. However, it is difficult to realize the high-performance microLEDs on a plastic substrate owing to high growth temperature of the inorganic-based LED layers (over 500 °C) as a critical thermal limit [86,88,89]. Fig. 4a illustrated AlGaInP flexible vertical microLEDs (f-VLEDs) fabricated by using an anisotropic conductive film (ACF)-based transfer and a backside chemical etching.[92] The ACF packaging was performed by the thermo-compression bonding (low temperature of 190 °C with uniaxial pressure of ~ 3 MPa), enabling the microLEDs to be simultaneously transferred and interconnected to a flexible printed circuit board (FPCB). The metal-coated polymer balls in the ACF resin were deformed by the concentrated pressure, sequentially forming the short current path between electrodes and microLED chips (Fig. 4b). Fig. 4c presents the developed ACF packaged f-VLED device that emits high-power red light ($25\,mW\,mm^{-2}$).

Furthermore, Lee et al. reported an ultrathin (thickness of 15 µm) and transparent GaN f-VLED array using a newly-developed monolithic fabrication process including an initial ILLO technique [93]. The GaN LED layers grown at a high-temperature condition were successfully transferred to a polymer substrate, overcoming the thermal limits of the plastic substrates. As shown in the Fig. 4d, the GaN f-VLEDs maintained its excellent optical and electrical properties after the ILLO process. All the high-density 30 × 30 blue f-VLEDs were well operated, which exhibited the outstanding irradiance of $30\,mW\,mm^{-2}$ at low forward voltage of ~2.5 V (Fig. 4e). As presented in Fig. 4f, high environmental durability of the monolithic f-VLED was confirmed by the harsh temperature-humidity test (105 °C and 85%), showing the expected lifetime of 11.9 years at room temperature.

2.2. Flexible optoelectronics for biomedical applications

Flexible microLEDs have been spotlighted as a promising candidate for self-powered biomedical tools due to its non-invasiveness, scalability and capability of multiple modulation and localized optical

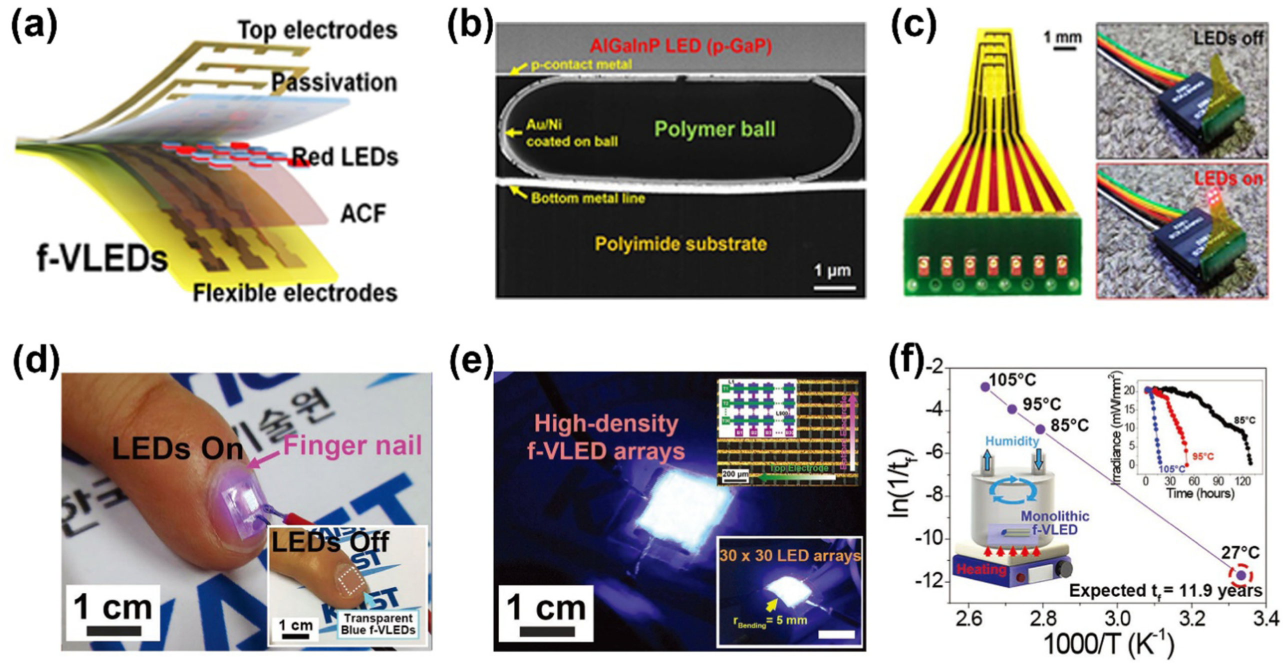

Fig. 4. (a) Schematic illustration of the ACF packaged AlGaInP f-VLEDs. (b) Cross-sectional SEM image of the thermos-compressed ACF, connecting the microLED chip to the polyimide substrate. (c) A photograph of the 4 × 3 ACF f-VLEDs, which are combined with the wired connector the printed circuit board. Reproduced with permission from [92]. Copyright 2018, Elsevier. (d) A photograph of the 30 × 30 blue f-VLED array. The upper inset is a magnified image of the passive-matrix GaN LED array. The lower inset is a picture of f-VLED on a metal stick with bending curvature radius of 5 mm. (e) A photograph of ultrathin transparent blue f-VLEDs on a human finger nail. The inset image show the f-VLEDs in an off-state. (f) Arrhenius plots of blue f-VLEDs to evaluate the lifetime of the device. The inset graphs exhibits the results of the irradiance consecutive measurement at 85, 95, and 105 °C. Reproduced with permission from [93]. Copyright 2018, John Wiley and Sons.

stimulation [5,94–98]. Several research teams have demonstrated various phototherapeutic systems including brain photostimulators, real-time monitoring devices and skin therapeutic apparatus [83,96–99]. Recently, Lee et al. reported the optogenetic stimulator using AlGaInP f-VLEDs for controlling body movements of a living mouse.[92] The red f-VLEDs were fabricated by the low-temperature ACF packaging process on the plastics, and carefully inserted under the mouse skull through a cranial slit. The AlGaInP f-VLEDs with a wavelength of 638 nm could activate the red-light driven channel rhodopsin (Chrimson), successfully stimulating the motor neurons of the mouse, as schematically illustrated in Fig. 5a. The movements of the mouse forelimb and whisker were analyzed by detecting the change of the electromyogram (EMG) signals during the red-light irradiation (Fig. 5b). According to the observation, the photo-stimulation of red f-VLED enabled the modulation of the muscular and neuronal behaviors without any unintended side effects.

Fig. 5c presents the schematic illustration of wearable pulse oximetry system on a silicone elastomer substrate, which was composed of the near-field communication (NFC) antenna, the Si photodiode and the packaged red/infrared (IR) LEDs [100]. As shown in Fig. 5d, the flexible oximeter device was wirelessly operated on the earlobe by the NFC power transfer. Heart rate (HR) and blood oxygenation (SpO_2) level could be acquired by analyzing the optical signal of red/IR LED after 70 s breath-hold test, as presented in Fig. 5e.

Lee et al. demonstrated a wearable trichogenic photostimulator using AlGaInP f-VLED arrays (Fig. 5f) [101]. By overcoming the processing thermal limits of plastics, high-performance red microLEDs on a flexible plastic substrates presented an irradiance of 30 mW mm^{-2}. The f-VLEDs were stably attached on the human skin, irradiating the red light without any thermal damages of the skin. The flexible photostimulator was utilized to promote the hair-regrowth of the depilated mice by irradiating red light (wavelength of 650 nm) during 20 consecutive days. Fig. 5g shows photographs of the hair-growth test results, which exhibits a localized hair-regrowth area resulting from the phototreatment. The light-exposed mice showed a fast hair-growth tendency in the shaved region compared to other control groups (i.e., non-treated and hair restorer-treated groups).

3. Flexible electronics for large-scale integration

3.1. Si-based f-LSI devices

f-LSI (e.g., flexible processors, memories and communication modules) that consists of more than thousands of highly integrated nano-devices have been considered as essential elements for self-powered flexible electronic systems since they perform core functional operations such as signal processing and data storage [102,103]. There have been various studies to realize f-LSIs especially based on single-crystalline Si to take advantage of its outstanding semiconducting characteristics (e.g., moderate bandgap and carrier mobility), thermal stability and compatibility to the standard CMOS processes [30,57,60,66,102,104,105]. However, critical restrictions exist in the Si-based f-LSIs processing on plastic substrates due to their thermal instability as well as multi-alignment inaccuracy [103,106]. Therefore, some researchers have attempted to transfer the high-performance LSIs constructed on conventional Si wafers onto flexible substrates using the backside chemical etching [59,60,104] or the mechanical exfoliation technology [56,66]. In 2013, Hwang et al. demonstrated Si-based f-LSI, which was 0.18 μm CMOS-processed flexible radio-frequency integrated circuits (RFICs), as shown in Fig. 6a [104]. Ultrathin (thickness of 145 nm) RFICs consisting of 1000 nano-transistors could be transferred on a plastic substrate by the backside etching of the handle silicon-on-insulator (SOI) wafer, showing excellent electrical properties (low insertion loss (< 1 dB) and high isolation (> 26 dB) at on and off states, respectively) and good flexibility. In 2015, Kim et al. reported ultrathin Si-based flexible NAND (f-NAND) flash memory interconnected on FPCB by bonding the device on FPCB through ACF packaging and subsequently removing the handle wafer via chemical etching, as presented in Fig. 6b [59]. The f-NAND flash memory retained large string current ratio over 20 without any degradation

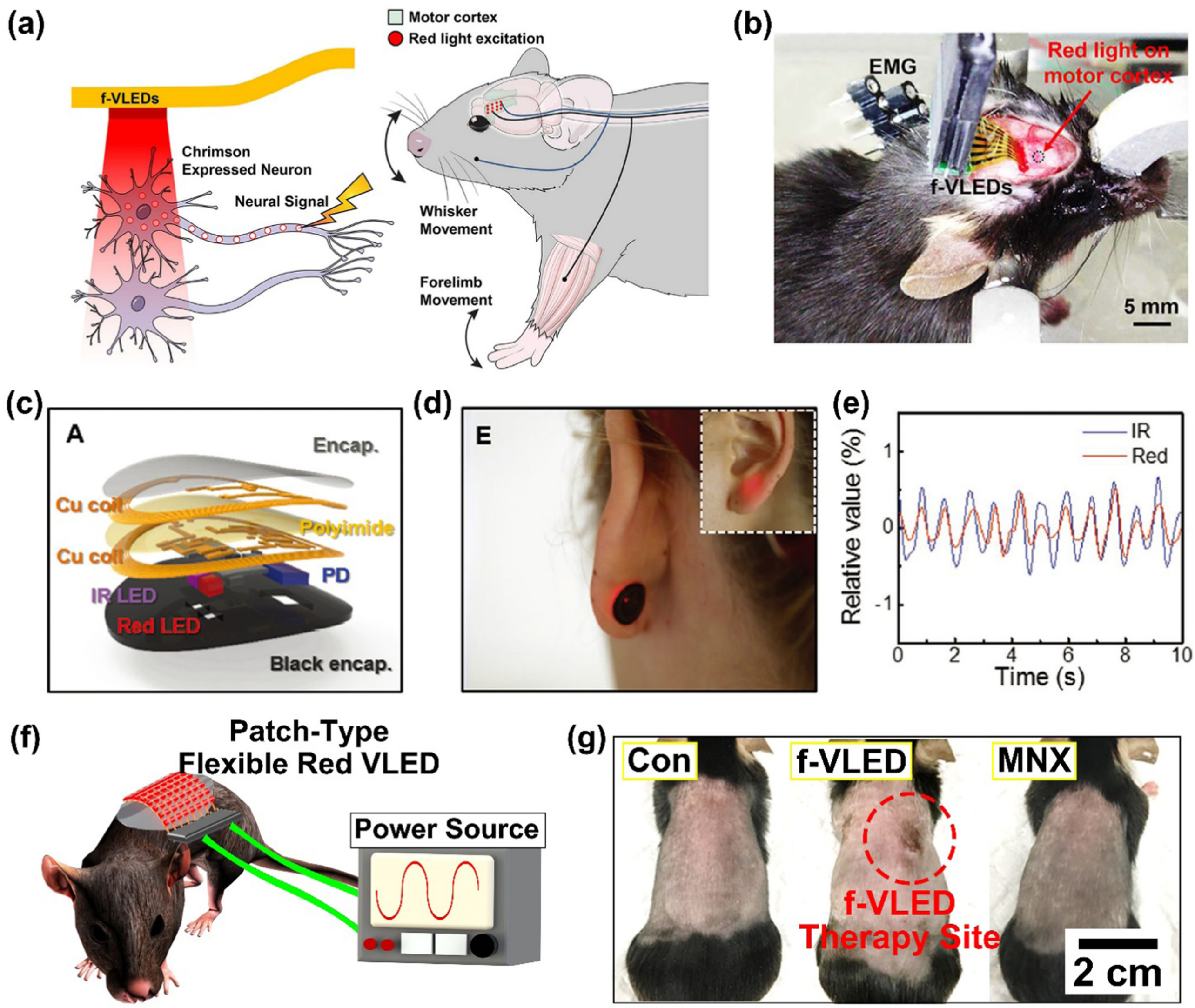

Fig. 5. (a) Schematic for the optogenetic stimulation of the mouse brain by red f-VLEDs. (b) A photograph of red f-VLEDs inserted under the mouse skull, stimulating the motor cortex. Reproduced with permission from [92]. Copyright 2018, Elsevier. (c) Schematic of the near-field communication (NFC) electronic systems, consisting of NFC antennas, red LEDs, infrared LEDs and photodiodes. (d) A photograph of wireless pulse oximeter attaching on the human earlobe. (e) Relative value graphs of red and IR signal acquired from the human earlobe. Reproduced with permission from [100]. Copyright 2017, John Wiley and Sons. (f) Schematic illustration of the hair-growth photostimulation using patch-type red f-VLEDs. (g) Photographs of hair-regrowth area in the mice dorsal skin after trichogenic experiments. Reproduced with permission from [101]. Copyright 2018, American Chemical Society.

during 1000 bending cycles, which was ascribed to the high flexibility of ultrathin f-NAND and the robust durability of ACF interconnection against bending deformation.

In addition, Kim et al. developed a continuous roll-based flexible ACF packaging method by simultaneously transferring and interconnecting the Si-based f-NAND flash memory array (16 × 16) during a highly-productive roll-to-plate process (Fig. 6c) [60]. Fig. 6d schematically illustrates the roll-based thermo-compression ACF bonding and backside etching method. As an initial step, the NAND flash memory was fabricated on a SOI wafer using conventional CMOS semiconductor processes. After adhering the device on a rigid intermediate glass substrate, the SOI handle wafer was gently eliminated in potassium hydroxide (KOH) wet-etching solution. By the roll-based ACF interconnection, the intermediate glass substrate was subsequently released, finalizing the high-performance f-NAND flash memory array. The f-NAND flash memory exhibited stable operation and reliable data-storage capability (10^2 of I_{on}/I_{off}, $\sim 10^3$ cycles of endurance, $\sim 10^4$ s of retention) even under repetitive bending deformation. As another approach for Si-based f-LSIs, a mechanical exfoliation method was employed to delaminate the upper layer of the LSI devices fabricated by the CMOS process by applying the principle of tensile surface stressor. [56,66] Tensile-stressed Ni film was deposited or electroplated to the LSI-included Si wafer. Then, it induces a high stress field beneath the wafer surface, which causes crack initiation and propagation that exfoliates the few tens of microns of the LSI-included top part of the wafer. Through this stress-based delamination technique, so called 'controlled spalling', Shahjerdi et al. demonstrated nanoscale f-LSIs including functional ring oscillators (RO) and memory cells, as shown in Fig. 6e [66].

3.2. Novel electronics for f-LSI

There have been efforts to demonstrate novel devices of f-LSI, which are based on various materials such as oxides, chalcogenides and ferroelectrics due to their outstanding switching characteristics and scalability [47,107–116]. Kim et al. utilized the ILLO technology to transfer a crossbar-structured one selector-one resistive switching memory (1S-1R) array from a rigid glass substrate to a flexible plastic sheet (Fig. 7a).

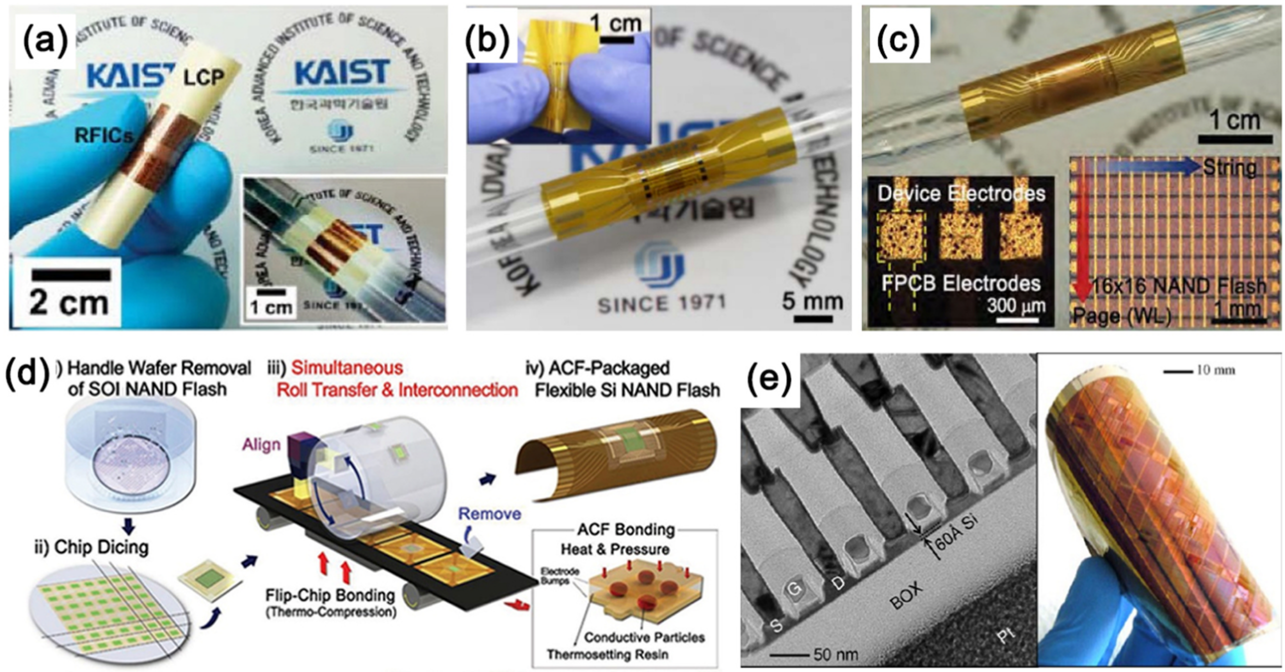

Fig. 6. (a) Photograph of the ultrathin Si-based flexible RFICs realized on a LCP substrate. The inset shows the device forming a conformal contact on curvilinear surfaces of three glass pipets. Reproduced with permission from [104]. Copyright 2013, American Chemical Society. (b) Image of the ACF-packaged flexible Si-based NAND flash memory wrapped on a glass rod. The inset presents the highly-flexible nature of the Si-based f-NAND memory during repetitive bending. Reproduced with permission from [59]. Copyright 2015, IEEE. (c) Photograph of the ultrathin Si-based f-NAND interconnected on FPCB realized by continuous roll-based ACF packaging process. The left inset is the magnified OM image of the electrode area showing the precise alignment between the device and the FPCB electrodes. The right inset shows the active memory area of the f-NAND without any degradations occurred during roll-based ACF packaging. (d) Schematic illustration describing the continuous roll-based ACF packaging process. Reproduced with permission from [60]. Copyright 2016, John Wiley and Sons. (e) (Left) Cross-sectional scanning electron microscopy (SEM) image of the demonstrated flexible nanoscale ultrathin n-FET. (Right) Photograph of the Si-based f-LSI realized by controlled spalling technique. Reproduced with permission from [66]. Copyright 2012, American Chemical Society.

[47] As shown in Fig. 7b, the established ILLO process enabled a 32 × 32 crossbar-array of integrated 1S-1R cells on a 50-μm-thick PET film without any degradation of their electrical and mechanical properties. Fig. 7c illustrates the fabrication process of flexible phase-change random access memory (f-PRAM) using a physical lift-off (PLO) technique [117]. A 32 × 32 parallel array of one conductive-filamentary phase-change material (PCM) and one Schottky diode for the PRAM device was fabricated on the Mo-based exfoliation layer. The ultrathin layer of the PRAM was physically exfoliated from the rigid glass substrate by the low adhesive binding energy of FCC/BCC Mo interface with the SiO_2 buffer layer, and then subsequently transferred onto a 12.5 μm-thick PET substrate to realize the f-PRAM array without thermal limits of plastic substrates. As presented in the photograph of Fig. 7d, the f-PRAM could be successfully detached from a glass substrate, indicating its outstanding flexibility. The flexible memory exhibited reliable memory operations without any mechanical degradation (e.g., cracks or wrinkles) during repeated bending fatigue tests. The developed PLO mechanism was deeply studied by first-principles density-functional theory (DFT) calculations, verifying the Mo phase transformation-based exfoliation method. Although the main phase of Mo is BCC in ambient environment, the FCC Mo phase becomes more dynamically stable than the BCC phase under the high tensile strain along uniaxial direction. Note that the sequentially deposited SiO_2 layer induces the uniaxial tensile stress at the interface with Mo layer due to the lattice mismatch. Therefore, the phase transformation of Mo occurs from the BCC to FCC under the strain [118]. Simultaneously, the FCC/BCC Mo interface exhibits lower binding energy compared to the pure FCC and BCC phase at the tensile strain condition. Therefore, Kim et al. concluded that the thermodynamically strain-induced Mo phase transition results in the fracture initiation and exfoliation, leading to the additional surface creation for the final PLO process [119,120]. It should be noted that this research not only performed experimental studies but also investigated the fundamental fracture mechanism of crack initiation and propagation to establish a physical delamination technology for controlling the transfer of ultrathin f-LSI [119,121,122].

4. Self-powered system on plastic substrates

4.1. Optimization of energy harvesting and self-powered system

We should utilize electrical energy to operate any electronic devices mentioned above all systems. In the modern society, portable and mobile energy devices have become important more and more because there are various electronics in many ubiquitous and convenient systems in our life [123]. In particular, energy harvesting devices have drawn attentions to support energy storage devices, using alternative sources from our environments[6,124]. Although solar energy (i.e., photovoltaic devices) has been developed in various levels, they have still significant disadvantages in some isolated spaces such as in vivo biological body and dark infrastructural environment [49,125]. Recently, mechanical energy harvesting technologies have been widely studied to optimize energy conversion from mechanical deformations to electrical energies in small-scale, portable, and device levels toward self-powered electronic devices [123,124]. This key trend is especially essential for flexible applications because the future electronics are going to flexible electronics. Moreover, flexing and bending motions of biological and machinery systems can be directly used as the mechanical energy harvesting sources [6].

Even though many researchers have studied a variety of mechanical energy harvesters, there are still critical limitations for practical use in real electronic systems. To achieve more useful energy harvesting applications, more systematic research flow should be established in an

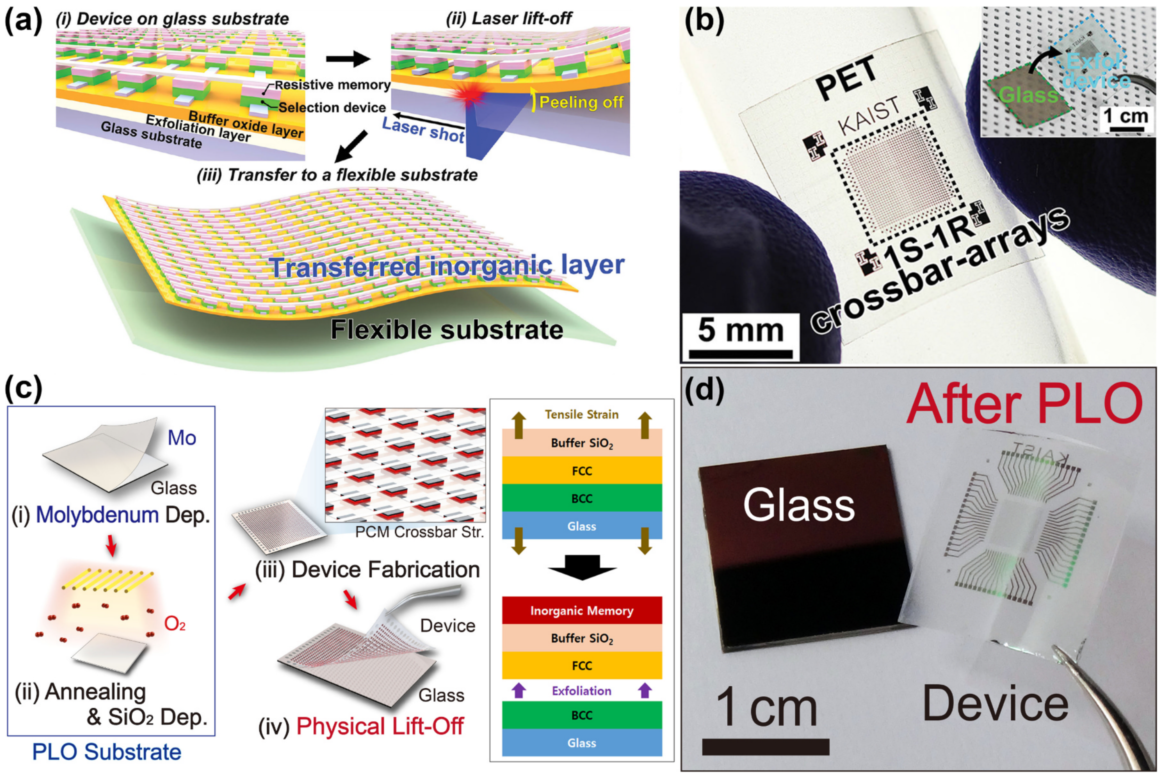

Fig. 7. (a) Schematic illustration showing the ILLO process for flexible RRAM fabrication. (b) Optical image of the ultrathin flexible 1 S-1 R RRAM device on PET substrate. Reproduced with permission from [47]. Copyright 2014, John Wiley and Sons. (c) Fabrication procedure of the f-PRAM fabrication process using the PLO transfer method. The right inset depicts the PLO mechanism, which is based on the Mo phase transformation. The left inset presents the device structure and the materials of the f-PRAM unit cell (i.e., vertically stacked 1 SD-1 CFPCM cell). (d) Photograph of the f-PRAM detached from a glass substrate.

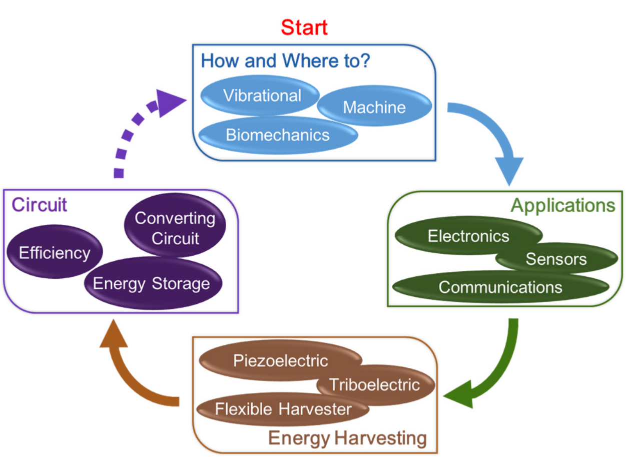

Fig. 8. Rational and logical research flow to realize practical energy harvesting and self-powered electronic system.

overall plan according to each device case. In this review part, we will point out the optimum engineering flow for practical energy harvesting technologies of self-powered system and some examples.

First of all, one needs to define the location and environment where the mechanical energy harvester is used. Because there are many types of mechanical energy around us (the top panel of Fig. 8), we should clarify the mechanical resources for device designs. Particularly, this starting stage is highly important in flexible devices due to its sensitively modified properties by materials and device structures. In the vibrational energy sources, for example, the energy harvesting devices should be fabricated with optimum thickness, mechanical modulus, mass tip, etc. to satisfy resonance-related behaviors, proper stiffness, and so on [127]. The device design is also affected by the type of applications. As pointed by the right panel of Fig. 8, the device purpose should be considered to set up the design such as area, size, encapsulation, etc [126]. In addition, the application type is directly associated with electrical and circuital characteristics which should be compatible to the whole design of self-powered system.

Next, one should select an energy harvesting principle and fabricate an actual device. There are two representative mechanical energy harvesting principles: piezoelectric and triboelectric (the bottom panel of Fig. 8). The piezoelectric principle, which is an electromechanical coupling phenomenon by intrinsic dipole displacement or rotation in crystallographic non-centrosymmetric materials, has been well known for past many decades, and studied for device-level generators recently. In contrast, the triboelectric principle, which is induced by charge transfers between two different arbitrary materials through macroscopic or microscopic frictions, has been demonstrated to invent new energy harvesting for the past five years. Although the triboelectric energy harvesters and nanogenerators can show very high power performance, it has generally some critical limitations such as the weak resistance to humid and abrasion, as well as the difficulty for flexible configuration with efficient friction [127,128]. We focus on piezoelectric energy harvesters because this review paper does deal with flexible electronics for future electronic systems.

To accomplish high-performance flexible piezoelectric energy harvesters, one need to realize high-quality piezoelectric ceramic films on flexible plastic substrate [6]. Even though some decent polymer or soft composite-based piezoelectric devices have been reported, [129–137]. piezoelectric ceramic materials still present much higher intrinsic piezoelectric properties. However, it is difficult to realize high-quality piezoelectric ceramic films on flexible plastic substrates because piezoceramics should be crystallized and annealed at high temperature (around 1000 °C), where plastic substrates cannot endure the processing. To overcome the serious thermal limits, the ILLO or the physical

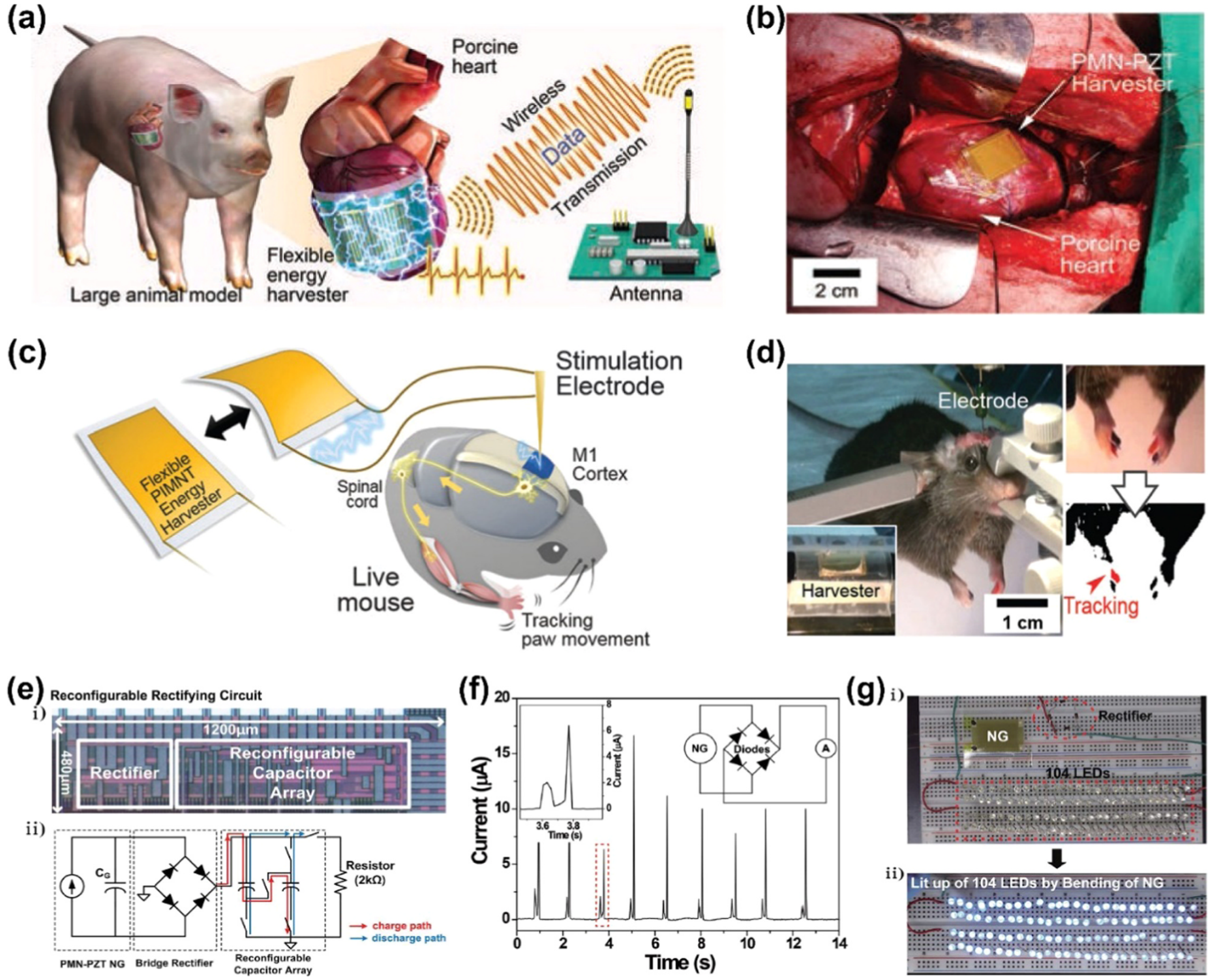

Fig. 9. (a) Schematic illustration of the in vivo self-powered wireless data transmission system. (b) The flexible film energy harvester is fixed on a porcine heart and generates electrical energy from periodical cardiac biomedical movement. Reproduced with permission from [14]. Copyright 2017, John Wiley and Sons. (c) Scheme for the animal experiment about the brain stimulation of mouse using the flexible piezoelectric energy harvester. The paw motion was tracked during the stimulation. (d) Photographs of a head-fixed mouse and the flexible piezoelectric energy harvesting stimulator (left panel). The flexible stimulator is connected to a bipolar electrode put in the M1 cortex. The movie-captured image was adjusted to binary images to track the paw movement of stimulated mouse (right panel). Reproduced with permission from [18]. Copyright 2015, Royal Society of Chemistry. (e) Top-view optical image (i) and schematic circuit diagram (ii) of the rectified reconfigurable conversion circuit. (f) Short-circuit current signals of the flexible piezoelectric energy harvester rectified by the reconfigurable circuit. (g) LED bulbs directly turned on by the flexible energy harvester through the reconfigurable circuit. Reproduced with permission from [4]. Copyright 2015, John Wiley and Sons.

exfoliation processes can be also efficiently used for the piezoelectric ceramic films and device structures, as introduced by the previous Sections [4,5,14,18,20,49,64,76,125]. Therefore, one can fabricate high-quality piezoelectric ceramic films and high-performance energy harvesters on the flexible plastic substrates.

Lastly, one should also investigate some extrinsic circuits connected with the energy harvesting devices, as presented in the left panel of Fig. 8. There are three critical issues about the generated electricity from energy harvesting devices, not only piezoelectrics but also triboelectrics; i) the energy harvesters produce electrical signals by mechanical input, but the output is generally shown as alternate, discrete peaks and irregular amplitudes in usual environments because most of mechanical inputs in our surroundings are irregular, arbitrary, and complicated. ii) Inherently, the generated charge flow and current density cannot be as high as the direct current (DC) of solar cells, whereas the generated voltage is very high, i.e., hundreds V level. iii) Although the generated power can be enhanced by materials and device structures, the efficiency can easily decrease through the external circuits due to various loss factors within the practical electronic and storage applications (e.g., dielectric, thermal, magnetic, etc.). Therefore, the circuit design is highly important to control conversion efficiency, energy loss, and real device operation for achieving practical uses of energy harvesting and self-powered systems [4,14,20].

4.2. Flexible self-powered energy harvesting system

According to the optimum research flow of energy harvesting devices, some systematic self-powered electronics have been studied for the past few years. Recently, Kim et al. developed a self-powered wireless data transmission using an in vivo biomechanical energy harvester [14]. Fig. 9a shows the schematic flow for the in vivo self-powered system with wireless devices. The researchers selected an in vivo environment of a large animal model for the biomedical energy harvesting. Additionally, the application was wireless transmission demonstrating the communication of vital signals between a biological system and an external antenna. To use heart beat biomechanical energy, the energy harvester was fabricated by high-quality single-crystalline piezoceramic film on a flexible plastic substrate using physical

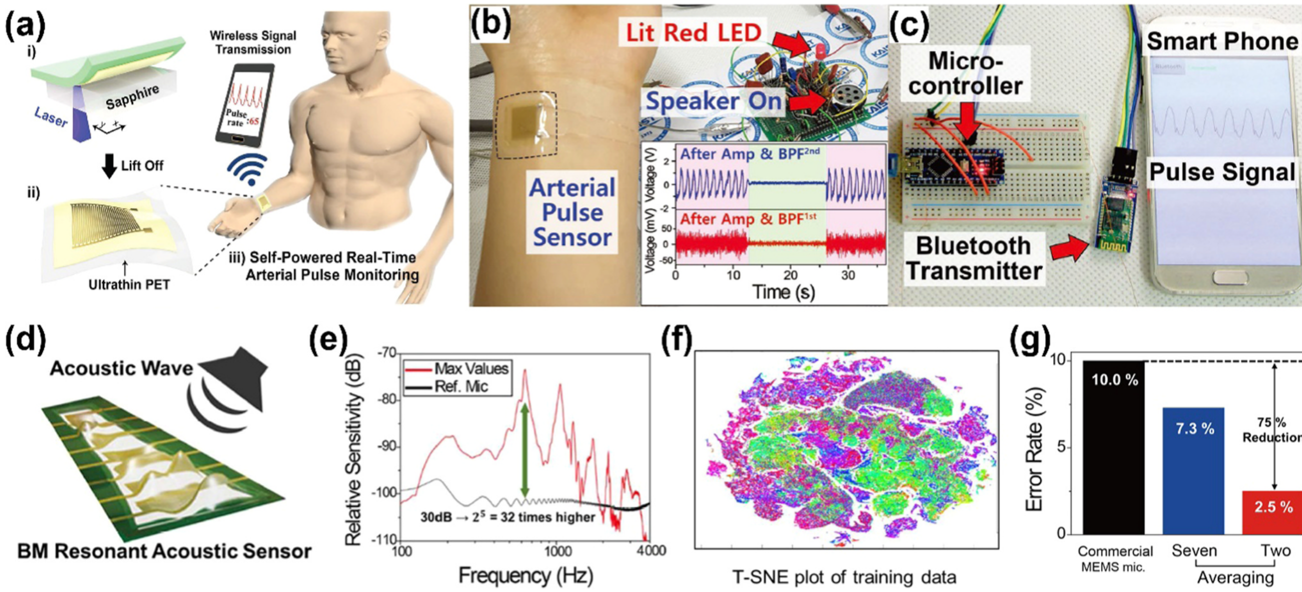

Fig. 10. (a) Schematic illustration of the fabrication process for self-powered pressure sensor: i) PZT thin film (2 µm) deposited on a sapphire wafer, supported by a thermal release tape, exfoliated via the ILLO process. ii) Transferred onto an ultrathin PET substrate (4.8 µm), thermal release tape is removed, and Cr and Au are deposited for IDEs. iii) Conformal contact of sensor on human wrist, and wireless transmission of detected pulse signals to a smart phone. (b) Photograph of the LED and speaker unit conducted synchronously according to the artery pulse. The inset indicates the output voltage from the first (bottom, red) and second (top, blue) amplifier stage. (c) Photograph of wireless transmission of the pulse to a smart phone, showing capability for a real-time arterial pulse monitoring system. Reproduced with permission from [19]. Copyright 2017, John Wiley and Sons. (d) Schematic illustration of the multi-channel f-PAS inspired by the resonant structure of the basilar membrane. (e) Relative sensitivity of the f-PAS and reference microphone over a voice frequency range from 100 to 4000 Hz. Reproduced with permission from [140]. Copyright 2018, Elsevier. (f) The trained STFT features visualized in t-SNE plot by 2800 training data of 40 people. The t-SNE plot embeds high-dimensional data of similar objects into a low-dimensional space, which is related with probability distribution. (g) Recognition error rate of commercialized phone and the f-PAS (seven-averaging and two-averaging) in the mixture number of 30. Reproduced with permission from [141]. Copyright 2018, Elsevier.

exfoliation processes overcoming the thermal limit related to the single-crystalline material (Fig. 9b). Finally, the wireless data transmission was successfully performed through the efficient capacitor-related conversion circuit and antenna design.

The similar optimized self-powered system was also demonstrated for brain stimulation using a high-performance flexible energy harvester by Hwang et al. [18]. Fig. 9c illustrates the experimental schematics for the stimulation of the specific brain part using the flexible piezoelectric energy harvester. The generated electrical signals by bending motions of the energy harvester are transferred to the biomedical interface electrode through a circuit (the left panel of Fig. 9d). To define the results of brain stimulation, the neural behavior of the mouse was investigated in detail by motion tracking system (the right panel of Fig. 9d). This novel approach could be more expanded to the biomedical implanted systems inside restricted space of biological surroundings, and generate electrical energy from biomechanical energy of organs such as lung, diaphragm as well as heart to support self-powered biomedical devices.

Sometimes, piezoelectric energy harvesting research should more concentrate on the circuit investigation, rather than materials or devices. Hwang et al. developed a reconfigurable power management circuit fabricated by the 0.25 µm CMOS process, [4] as shown in the optical microscopy image of Fig. 9e. The simplified circuit diagram presents the specific connections of a flexible energy harvester, a reconfigurable capacitor charger array, a low-loss rectifier, and an external load resistor. Note that the array of multiple capacitors can modify the amount of capacitance by adjusting serial and parallel arrangements of capacitors. Therefore, the rectified maximum output current from the flexible energy harvester was about 16 µA which is 4 times higher electrical energy (~ 6.25 µJ) than that of a conventional rectifying circuit (Fig. 9f). Using the optimized circuit and the thermal limit-overcoming flexible energy harvester, 104 blue LED bulbs were brightly turned on without any other external power source, as presented by Fig. 9g.

Perovskite structured piezoceramics can be also utilized for achieving the pyroelectric energy harvesters which convert from temperature change to electricity due to pyroelectric and electrocaloric effects [138,139]. Using the state-of-an-art technologies overcoming thermal limits as described above, the high-performance flexible piezoelectric energy harvesters can be also modified for new flexible pyroelectric devices because most of perovskite piezoceramics also have pyroelectric properties.

Meanwhile, some piezoelectric polymer materials (i.e., polyvinylidene fluoride (PVDF) and relevant copolymers) have been also studied for a variety of piezoelectric energy harvesting systems because they can be processed in low temperature and show inherently soft for flexible applications [140–142]. However, PVDF has also critical drawbacks such as lower piezoelectric constants than those of perovskite structured piezoceramics.

Recently, the piezoelectric properties and applications of biomaterials (e.g., collagen, chitin, gelatin, cellulose, etc.) have drawn tremendous attentions to develop new biocompatible devices and unveil the principles of piezoelectric biomaterials [143–149]. It is very interesting since the piezoelectric effects of biomaterials should be addressed for bioelectricity and structural biology. Nonetheless, it must be noted that there are serious and significant controversies in the electromechanical properties of biomaterials due to other possible mechanisms [150]. Therefore, thorough fundamental studies as well as device developments should be conducted.

The triboelectric energy harvesting technology is another important field for mechanical energy harvesting. Triboelectric generators are performed by the mechanism of triboelectric effect of two different surfaces and electrostatic induction of connected circuits [151]. This new field has attracted many attentions from various researchers because the high performance and the simple device structure of triboelectric generators [127,128]. Moreover, most of triboelectric devices

Table 1
The main technologies for self-powered flexible electronic system beyond the thermal limit of plastic, and their mechanism.

Technology	**Light-materials Interaction**		**Optoelectronics**		**Self-powered system**		**Flexible electronics**		
Mechanism	Anneal	Synthesis	Laser Lift-Off	Chemical Etching	Laser Lift-Off	Physical Exfoliation	Laser Lift-Off	Chemical Etching	Physical Exfoliation
Material	BCP, AgNW, CuNP	Graphene, Metal-Oxide NW	GaN	AlGaInP, GaAs	PZT, Li-KNN	PMN-PT, PMN-PZT, PZT, PIMNT	a-Si:H	Si	Mo
Advantage	Fast process, High scalability		Fast process, High scalability	Low-cost process	Fast process, High scalability	Fast process	Fast process, High scalability	Low-cost process	Fast process
Disadvantage	Need of high-cost laser facility		Need of high-cost laser facility	Slow process	Need of high-cost laser facility	Low scalability	Need of high-cost laser facility	Slow process	Low scalability
Reference	[39,51,52,70,75]	[40,67,71,74]	[91,93,94,97]	[5,90,92,101]	[19,20,138–140]	[4,14,18,64]	[47,48,80]	[59,60,104]	[56,66,117]

have been made of polymer materials and organic films, which are mechanically flexible as well as low-temperature processible [151]. However, the performance of triboelectric devices can be easily degraded by external humidity and mechanical abrasion. Although these limitations have been improved by specific packaging or device structures [152], it could not be fully resolved such as complex architecture, mechanical inflexibility, and performance decrement. There are still various fundamental and engineering problems in the field of triboelectric devices [153].

4.3. Flexible self-powered sensor

Self-powered sensors have been spotlighted as a next-generation electronic system due to their considerable merits that do not require battery replacement, power supply wiring and external bias voltage [14,20,126,127,154,155]. Park et al. demonstrated a self-powered flexible pulse sensor based on the ILLO process for a real-time healthcare monitoring system, as schematically illustrated in Fig. 10a [19]. The piezoelectric PZT thin film was transferred to the ultrathin PET sheet of 4.8 µm thickness from the rigid substrate without thermal limits. As shown in Fig. 10b, the pulse sensor on the plastic substrate could be conformally adhered to a human wrist, providing the pulse signal in response to the arterial tension of human body. The red LED and speaker components were operated according to the piezoelectric output voltage generated by the artery pulse for a biomedical monitoring SoP. The radial artery pulse signal produced by the self-powered flexible pulse sensor was wirelessly transmitted to the smart phone using a micro controller unit (MCU, Arduino-Nano ATMEGA 168) and bluetooth transmitter board (Fig. 10c).

Han et al. reported a basilar membrane (BM)-inspired flexible piezoelectric acoustic sensor (f-PAS) based on a multi-channel structure, as illustrated in Fig. 10d [156]. The high-performance PZT on the sapphire substrate was transferred to a thin plastic substrate using the ILLO process to overcome thermal limits of the plastics. The flexible PZT film was shaped as a concaved trapezoidal design, and then subsequently integrated by multiple interdigitated electrodes (IDEs). The multichannel f-PAS successfully covered the human voice frequency band (from 100 Hz to 4000 Hz), exhibiting 32 times higher maximum relative sensitivity (-76 dB at 650 Hz) compared to a reference condenser microphone (Fig. 10e).

Han et al. [157] also developed a machine learning-based speaker recognition SoP using the self-powered f-PAS. The f-PAS acquired the abundant and intrinsic voice information from a single sound input through the highly sensitive multi-resonant PZT membrane. The machine learning based Gaussian Mixture Model (GMM) algorithm was employed for identifying the speake [158]. The standard TIDIGITS datasets (20 men and 20 women speakers, 77 speeches per each speaker, total 3080 voice data) were recorded by the multi-channel f-PAS, which were converted to a frequency domain data by using a Fast Fourier Transform (FFT) and a Short-Time Fourier Transform (STFT). Among 3080 TIDIGITS voice data recorded by the f-PAS, the 2800 data were utilized for training step and the remaining 280 data were used for testing process to identify speakers. Fig. 10f presents the STFT features of the trained voice speeches visualized in a t-Distributed Stochastic Neighbor Embedding (t-SNE) plot, which is related to the probability distribution of similar clusters for the 2800 training data. The f-PAS showed 75% lower error rate than the commercialized microelectromechanical systems (MEMS) microphone by using the highest and the second highest electrical signals generated from the multi-channels of the f-PAS under acoustic stimulus in the voice frequency range (Fig. 10g). It should be noted that these flexible and sensitive self-powered sensors might be also applied by the principle of triboelectric devices because the triboelectric sensors have been well developed in pressure and touch sensors [152].

5. Conclusion

This review summarizes the novel approaches and technologies to achieve the self-powered flexible electronic systems based on inorganic materials by resolving the thermal limits. The inherently high-performance inorganic materials should follow inevitably high temperature processes for annealing or crystallization: These thermal limits hindered the highly-efficient flexible electronic devices. As shown in Table 1, recent novel approaches that enable the high-performance SoP include convergence of laser-material interactions, optoelectronics, self-powered energy devices, and large-scale integrated electronics by employing the newly developed technologies, i.e, laser-assisted methodologies, physical-mechanical exfoliation principles, or chemical treatments. Despite technical advances in overcoming thermal limits, several challenges still remained on the path to the commercialization of self-powered fully-flexible electronic system. For example, the exfoliation speed of functional components has to be significantly improved to satisfy industrial standards of self-powered SoPs. Another issue is an efficient integration of unit devices in terms of power consumption since energy harvesters have a limitation of matching electrical power supply from the system perspectives. We believe that these approaches become the main developing trend for a fully integrated self-powered flexible system. Self-powered electronic flexible electronic technologies beyond thermal limits of flexible substrates will shed light on unprecedented high-performance energy and biomedical applications for future sustainable technology and human society.

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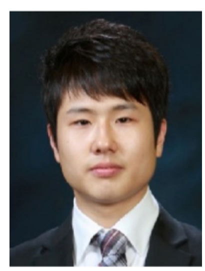

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