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# All-in-one energy harvesting and storage devices

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# All-in-one energy harvesting and storage devices

## **Abstract**

Currently, integration of energy harvesting and storage devices is considered to be one of the most important energy-related technologies due to the possibility of replacing batteries or at least extending the lifetime of a battery. This review aims to describe current progress in the various types of energy harvesters, hybrid energy harvesters, including multi-type energy harvesters with coupling of multiple energy sources, and hybridization of energy harvesters and energy storage devices for self-powered electronics. We summarize research on recent energy harvesters based on the piezoelectric, triboelectric, pyroelectric, thermoelectric, and photovoltaic effects. We also cover hybrid cell technologies to simultaneously generate electricity using multiple types of environmental energy, such as mechanical, thermal, and solar energy. Energy harvesters based on the coupling of multiple energy sources exhibit enhancement of power generation performance with synergetic effects. Finally, integration of energy harvesters and energy storage devices is introduced. In particular, self-charging power cells provide an innovative approach to the direct conversion of mechanical energy into electrochemical energy to decrease energy conversion loss.

## **Keywords**

all, one, harvesting, storage, energy, devices

## **Disciplines**

Engineering | Physical Sciences and Mathematics

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## All-in-one energy harvesting and storage devices

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Sang-Woo Kim<sup>\*ac</sup> and Jung Ho Kim<sup>\*b</sup>

Currently, integration of energy harvesting and storage devices is considered to be one of the most important energy-related technologies due to the possibility of replacing batteries or at least extending the lifetime of a battery. This review aims to describe current progress in the various types of energy harvesters, hybrid energy harvesters, including multi-type energy harvesters with coupling of multiple energy sources, and hybridization of energy harvesters and energy storage devices for self-powered electronics. We summarize research on recent energy harvesters based on the piezoelectric, triboelectric, pyroelectric, thermoelectric, and photovoltaic effects. We also cover hybrid cell technologies to simultaneously generate electricity using multiple types of environmental energy, such as mechanical, thermal, and solar energy. Energy harvesters based on the coupling of multiple energy sources exhibit enhancement of power generation performance with synergetic effects. Finally, integration of energy harvesters and energy storage devices is introduced. In particular, self-charging power cells provide an innovative approach to the direct conversion of mechanical energy into electrochemical energy to decrease energy conversion loss.

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### 1. Introduction

Exploring the possibilities of renewable, sustainable, green energy sources to replace fossil fuels is one of the most significant and challenging issues in energy research because of air/water pollution and oil depletion due to the use of fossil fuels. In particular, renewable forms of energy such as sunlight, wind,



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rain, tides/waves, and geothermal heat have been extensively studied as alternative energy sources to end the age of oil.<sup>1–5</sup> Eventually, these forms of energy must be integrated with the major power grids to meet the requirements of megawatt to gigawatt power scales.

In recent years, smart electronics with multiple functionalities such as tablets, mobile phones, and various sensors have become widespread everywhere in our daily lives. Even if such devices are designed for high energy efficiency and can run on a battery for a long time period, the power consumption is often heavily loaded because of the relatively large size of devices. In these circumstances, therefore, sustainable power sources are very much required for the independent, maintenance-free, and continuous operation of such low-energy-consumption smart electronics.

Over the past decades, energy harvesters based on interesting phenomena, such as the piezoelectric<sup>6–8</sup> and triboelectric<sup>9–11</sup> effects for harvesting mechanical energy, the pyroelectric<sup>12–14</sup> and thermoelectric<sup>15–17</sup> effects for harvesting thermal energy, and the photovoltaic (PV)<sup>18–20</sup> effect for harvesting solar energy, have been extensively studied for practical applications. These energy technologies are simply classified by their different energy conversion mechanisms, but the aim of all the energy harvesters is the conversion of wasted environmental energy to electricity. Nevertheless, all of the energy harvesters utilize only one type of energy, with the other types wasted. A PV cell, for example, is only designed to generate electricity under light illumination, and efficiency will be dramatically decreased under room light in indoor situations



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where sunlight is not available. Furthermore, thermal energy also appears in conjunction with mechanical and light energy. Thermal energy generated by mechanical and light energy will be wasted without a thermal energy harvester. Since micro/nanoscale smart systems can work under complicated environments and conditions, the use of only one type of energy harvester is insufficient to drive their operation. Thus, it is highly desirable to integrate energy harvesters so as to accumulate multiple types of energy for conversion into electricity, so that waste energy can be fully utilized and smart systems can be powered at any time and in any place.

Energy storage devices are just as important as energy harvesters in terms of energy technology today. This technology enables us to utilize energy to drive electronic devices/systems. Therefore, the integration of energy harvesting and storage devices (*i.e.* all in one) is the ultimate purpose following the rapid development of devices and technology, which offer a possible solution. This is a newly rising field in energy research, relating to the applications of nanomaterials and nanotechnology for harvesting energy to power micro/nano-systems. It can be used to possibly replace batteries or at least extend the lifetime of batteries.

Herein, we introduce recent progress in research on the integration of energy harvesting and storage devices. First, different energy harvesters are categorized by their energy sources, such as mechanical energy, thermal energy, and solar energy, and their operating mechanisms, such as the piezoelectric, triboelectric, pyroelectric, thermoelectric, and PV effects. Second, research on various integrated multi-type energy harvesters is summarized, and enhancement of their output performance by the synergetic coupling effect of multiple energy sources is also described. Third, the integration of energy harvesters and energy storage devices is summarized, including simple integration and hybrid structures in a single cell. Finally, this review will give more practical guidelines for overcoming the current obstacles.

## 2. Energy harvesting

### 2.1 Mechanical energy harvesting

**2.1.1 Piezoelectric effect.** Mechanical energy such as stress/strain can be converted into electrical energy by the piezoelectric effect (Fig. 1a). This stress/strain can come from many different sources that exist everywhere, such as vibrations, body motion, acoustic noise, and air flow. The piezoelectric effect is known as a coupling phenomenon between mechanical and electrical states. When mechanical stress (or strain) is applied to a piezoelectric material, the crystal structure of the material is deformed, and this causes movement of electrical charges. The polarization charge density due to the electrical moment is proportional to the applied mechanical stress, which is given by eqn (1):

$$\rho = dX \quad (1)$$

where  $\rho$  is the polarization charge density,  $d$  is the piezoelectric coefficient, and  $X$  is the applied stress. Then, the charge density results in electric field and potential as follows, eqn (2):

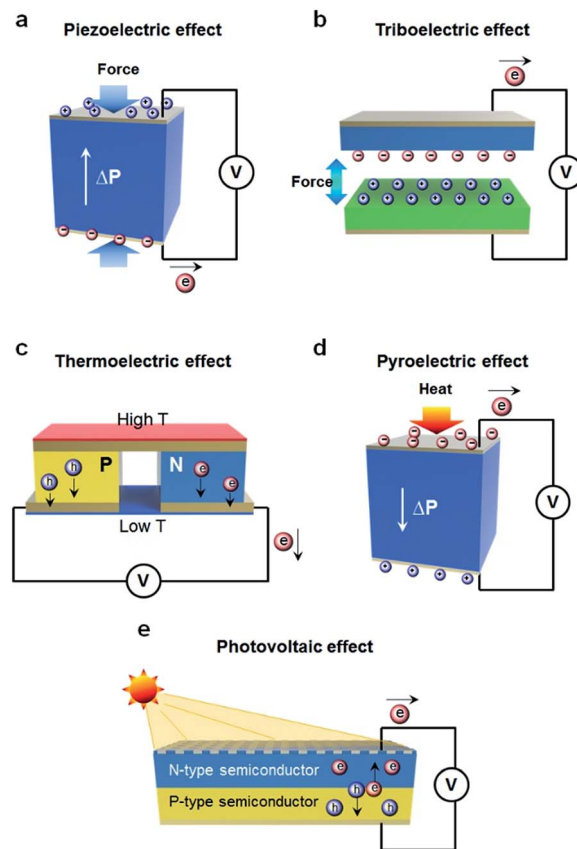


Fig. 1 Schematic illustration of energy harvesters based on (a) the piezoelectric effect, (b) the triboelectric effect, (c) the thermoelectric effect, (d) the pyroelectric effect and (e) the PV effect.

$$\nabla E = \frac{\rho}{\epsilon} \quad (2)$$

where  $\nabla E$  is the divergence of the electric field,  $\rho$  is the charge density, and  $\epsilon$  is the permittivity. Therefore, by designing a device that comprises a piezoelectric material, electrodes, and an external circuit, the electric potential due to the piezoelectric effect can generate electric current and can be used as a source of electrical energy. The so-called piezoelectric nanogenerator (PENG) was first invented in 2006 by Prof. Wang in the Georgia Institute of Technology by sweeping an atomic force microscope (AFM) tip across a vertically grown ZnO nanowire.<sup>6</sup> Thenceforth, the PENG research field has received great attention, and various types of PENGs have been reported. Vertically grown piezoelectric nanostructure based PENGs,<sup>21–26</sup> laterally aligned piezoelectric nanostructure based PENGs,<sup>27–31</sup> stretching type PENGs,<sup>32–34</sup> mechanically durable nanocomposite based PENGs,<sup>35–39</sup> and highly sensitive nano-thin film-based PENGs<sup>40–43</sup> were investigated. In addition, various semiconducting piezoelectric nanostructures in materials such as ZnO,<sup>44,45</sup> GaN,<sup>46</sup> CdS,<sup>47,48</sup> InN,<sup>49</sup> and insulating piezoelectric materials such as PbZrTiO<sub>3</sub>,<sup>50,51</sup> BaTiO<sub>3</sub>,<sup>53,54</sup> KNbO<sub>3</sub>,<sup>55</sup> NaNbO<sub>3</sub>,<sup>56</sup> and poly(vinylidene fluoride)-co-trifluoroethylene [P(VDF-TrFE)]<sup>31,42,57</sup> have been intensively applied to fabricate PENGs. These fabricated PENGs are subjected to various forms of external mechanical energy such as vibrations, sound,<sup>43</sup> rain drops,<sup>57</sup> pressing,<sup>57</sup> bending,<sup>51</sup>

stretching,<sup>32</sup> heart beats,<sup>52</sup> muscle movements,<sup>52</sup> inhalation,<sup>52</sup> and wind<sup>43</sup> to generate electrical energy.

**2.1.2 Triboelectric effect.** Newly invented triboelectric nanogenerators (NGs) (TENGs), based on the well-known contact electrification effect, provide a new approach to generating electricity from mechanical energy to operate small electronic devices. In 2012, TENGs were invented and explained on the basis of electrostatic and contact electrification physics (Fig. 1b).<sup>9</sup> In the contact electrification effect, a material surface becomes electrically charged after it comes into contact with a different material through friction, owing to charge transfer between the two materials. These transferred charges remain for a long time on their respective surfaces. An electrostatically charged material causes a potential, and it drives induced electrons to flow between the electrodes by periodic contact and separation of the two materials. The generated electric potential  $V$  can be calculated using eqn (3):

$$V = -\frac{\rho d}{\epsilon_0} \quad (3)$$

where  $\rho$  is the triboelectric charge density,  $\epsilon_0$  is the vacuum permittivity, and  $d$  is the interlayer distance in a given state. The current  $I$  generated across an external load can be defined as follows, eqn (4):

$$I = C \frac{\partial V}{\partial t} + V \frac{\partial C}{\partial t} \quad (4)$$

where  $C$  denotes the capacitance of the system and  $V$  is the voltage across the two electrodes. The first term is the change in potential between the top and bottom electrodes due to the triboelectric charges. The second term is the variation in the capacitance of the system when the distance between two electrodes is changed due to the mechanical deformation. Numerous advantages of TENGs, including superior power output performance, many material options, easy tailoring of device structures, cost-effectiveness, the facile fabrication of large areas for applications, and stability and robustness, as well as environmental friendliness, will bring more opportunities to our daily lives in the near future.

Depending on the configuration of the electrodes and the different ways in which the triboelectric layers can be arranged, four operation modes of TENGs have been developed: the vertical contact mode, the lateral sliding mode, the single electrode mode, and the free-standing mode.<sup>58</sup> Their applications as various self-powered nanosystems, such as acceleration sensors, motion vector sensors,<sup>59</sup> biomedical monitoring systems,<sup>60</sup> electrochromic devices,<sup>61</sup> sound recording systems,<sup>62</sup> pressure sensors,<sup>63</sup> angle measurement sensors,<sup>64</sup> active tactile sensor systems,<sup>65</sup> tactile imaging devices,<sup>66</sup> electroluminescent systems,<sup>67</sup> and mercury-ion detection systems,<sup>68</sup> have been recently demonstrated. The triboelectric output can be further enhanced through control of electron affinity, as well as the work function, chemical structure, pressure, and surface roughness of the materials.

## 2.2 Thermal energy harvesting

**2.2.1 Thermoelectric effect.** There are two types of energy harvesters using thermal energy. One is based on the Seebeck

effect, which utilizes a temperature difference between the two ends of the device to drive the diffusion of charge carriers, which is called the thermoelectric (TE) effect (Fig. 1c). The thermoelectric effect, known since the 19<sup>th</sup> century, provides an interesting perspective for the conversion of heat to electrical energy. Given a thermal gradient, a thermoelectric generator (TEG) is able to convert heat into electrical energy even with small temperature differences. TEGs are simple, compact, robust, and very reliable because they contain no moving mechanical parts. For all these reasons, TEGs are attractive for a large variety of applications, in particular in the fields of green and renewable energy harvesting. The Seebeck effect can be expressed as eqn (5):

$$V = \alpha \Delta T \quad (5)$$

where  $V$  is the thermoelectric voltage,  $\Delta T$  is the temperature gradient, and  $\alpha$  is the Seebeck coefficient. Based on the thermoelectric effect, the efficiency of thermoelectric devices is determined by the thermoelectric material's figure of merit,  $ZT$ , which is a function of several transport coefficients as follows, eqn (6):

$$ZT = \frac{\sigma S^2 T}{\kappa_e + \kappa_l} \quad (6)$$

where  $\sigma$  is the electrical conductivity,  $S$  is the Seebeck coefficient,  $T$  is the mean operating temperature, and  $\kappa$  is the thermal conductivity. The subscripts e and l on  $\kappa$  signify the electronic and lattice contributions, respectively. A high figure of merit corresponds to high efficiency of the TEG. Therefore, there is serious interest in improving the figure of merit of thermoelectric materials for many industrial and energy applications. The most common material for thermoelectricity has been Bi<sub>2</sub>Te<sub>3</sub> since 1954, because its high electrical conductivity after doping and low thermal conductivity give a  $ZT$  of about 0.7–0.8 at room temperature.<sup>69–71</sup> Furthermore, various thermoelectric materials such as BiSbTe<sub>3</sub>,<sup>72–74</sup> PbTe,<sup>75,76</sup> CoSb<sub>3</sub>,<sup>77,78</sup> SiGe,<sup>79,80</sup> and Mg<sub>2</sub>Si<sup>81,82</sup> have been reported for TEGs.

**2.2.2 Pyroelectric effect.** The other energy harvesting technology using thermal energy is pyroelectric, based on the change in spontaneous polarization in certain anisotropic solids due to temperature fluctuation. Usually, harvesting thermal energy mainly relies on the Seebeck effect. When the temperature varies in a time-dependent way, however, without a spatial gradient, the Seebeck effect is not able to harvest thermal energy. In this case, the pyroelectric effect could be used to harvest waste thermal energy (Fig. 1d). The working mechanism of a pyroelectric nanogenerator (PNG) will be explained for two different cases: the primary pyroelectric effect and the secondary pyroelectric effect. The primary pyroelectric effect is related to the charge generation because of the change in polarization with temperature when the dimensions of the pyroelectric material are fixed. The secondary pyroelectric effect is an additional contribution of piezoelectrically induced charge by thermal expansion of a pyroelectric material with temperature change. The total pyroelectric effect is the sum of the primary and secondary pyroelectric effects. The pyroelectric coefficient,  $p$ , can be explained by eqn (7):

$$e = \frac{d\rho}{dT} \quad (7)$$

where  $e$  is the pyroelectric coefficient,  $\rho$  is the spontaneous polarization and  $T$  is the temperature. The electric current generated by the pyroelectric effect is expressed as eqn (8):

$$I = \frac{dQ}{dt} = \mu e A \frac{dT}{dt} \quad (8)$$

where  $Q$  is the induced charge,  $\mu$  is the absorption coefficient of radiation,  $A$  is the surface area, and  $dT/dt$  is the rate of temperature change. Therefore, when pyroelectric materials are heated or cooled ( $dT/dt > 0$ , or  $dT/dt < 0$ ), the overall polarization in the dipole moment is decreased or increased, which causes current to flow in the circuit. Usually, ferroelectric materials such as lead zirconate titanate (PZT),<sup>83,84</sup> BaTiO<sub>3</sub>,<sup>85</sup> P(VDF-TrFE),<sup>86–88</sup> and KNbO<sub>3</sub>,<sup>89</sup> and some piezoelectric materials which have spontaneous polarization, such as ZnO<sup>90</sup> and CdS,<sup>91</sup> are used to fabricate pyroelectric generators. By harvesting the waste thermal energy, pyroelectric nanogenerators have potential applications such as environmental monitoring, temperature imaging, medical diagnostics, and personal electronics.

### 2.3 Solar energy harvesting

Solar energy is regarded as one of the most important and realizable renewable energy sources due to its advantages of practically infinite quantity, a universal energy source, and cleanness.<sup>92</sup> Among the various solar technologies, such as solar heating, PVs, photosynthesis, *etc.*, the PV effect can convert light to usable electricity in the form of a voltage or electric current. A solar cell is an electrical device that converts the energy of light directly into electricity by using the PV effect,<sup>93,94</sup> which is related to both the chemical and physical phenomena and the materials (Fig. 1e). When light is incident upon a solar cell, electricity is generated through the light absorption by the semiconductor, excitation, hole/electron separation, and the transport of charges to the electrodes.<sup>93,94</sup> The conversion efficiency ( $\eta$ ) of a solar cell is the most commonly used parameter to define a cell's performance and compare it to that of another cell. The efficiency is defined as the ratio of energy output from the solar cell to input energy from the sun.<sup>94</sup> In addition to reflecting the performance of the solar cell itself, the efficiency depends on the spectrum and intensity of the incident sunlight and the temperature of the solar cell. Therefore, the conditions under which efficiency is measured must be carefully controlled in order to compare the performance of one device to another. Terrestrial solar cells are measured under AM 1.5 conditions and at a temperature of 25 °C. The efficiency of a solar cell is determined by the fraction of incident power which is converted to electricity and is defined as eqn (9):<sup>93,94</sup>

$$\eta(\%) = \frac{P_{\max}}{P_{\text{in}}} = \frac{V_{\text{oc}} \times J_{\text{sc}} \times \text{FF}}{P_{\text{in}}} a \times 100 \quad (9)$$

where  $V_{\text{oc}}$  is the open-circuit voltage,  $J_{\text{sc}}$  is the short-circuit current, and FF is the fill factor. The open-circuit voltage ( $V_{\text{oc}}$ ) is the maximum voltage available from a solar cell, and this occurs at zero current. The open-circuit voltage corresponds to the

amount of forward bias on the solar cell due to the bias of the solar cell junction with the light-generated current. The short-circuit current ( $J_{\text{sc}}$ ) is the current through the solar cell when the voltage across the solar cell is zero (*i.e.*, when the solar cell is short-circuited). The short-circuit current is equal to the absolute number of photons converted to hole–electron pairs due to the generation and collection of light-generated carriers. For an ideal solar cell with, at most, moderate resistive loss mechanisms, the short-circuit current and the light-generated current are identical. Therefore, the short-circuit current is the largest current which may be drawn from the solar cell. The short-circuit current and the open-circuit voltage are the maximum current and voltage, respectively, from a solar cell. At both of these operating points, however, the power from the solar cell is zero. The fill factor (FF) is a parameter which, in conjunction with  $V_{\text{oc}}$  and  $J_{\text{sc}}$ , determines the maximum power of a solar cell. The FF is defined as the ratio of the maximum power from the solar cell to the product of  $V_{\text{oc}}$  and  $J_{\text{sc}}$ . Graphically, the FF is a measure of the “squareness” of the solar cell and is also the area of the largest rectangle which will fit into the  $I$ - $V$  curve. As FF is a measure of the “squareness” of the  $I$ - $V$  curve, a solar cell with a higher voltage has a larger possible FF, since the “rounded” portion of the  $I$ - $V$  curve takes up less area. A commonly used expression for the FF can be determined empirically as eqn (10)<sup>93,94</sup>

$$\text{FF} = \frac{P_{\max}}{V_{\text{oc}} \times J_{\text{sc}}} = \frac{V_{\max} \times J_{\max}}{V_{\text{oc}} \times J_{\text{sc}}} \quad (10)$$

In general, solar cells can be sorted according to the semiconductor materials used for the active layer.<sup>92</sup> Typically, solar cells are classified as silicon-based solar cells,<sup>95,96</sup> dye-sensitized solar cells (DSSCs),<sup>97–100</sup> organic solar cells, including organic unit molecules<sup>101</sup> and polymers,<sup>102,103</sup> quantum-dot (QD) solar cells,<sup>104,105</sup> and perovskite solar cells.<sup>106–108</sup> In a specific cell, each of the factors  $V_{\text{oc}}$ ,  $J_{\text{sc}}$ , and FF is affected by the material species, cell structure, fabrication process, and other technologies.<sup>109–111</sup> To increase the cell efficiency by improving  $V_{\text{oc}}$ ,  $J_{\text{sc}}$ , and FF, therefore, many approaches are used, such as the development of novel materials,<sup>112–115</sup> light harvesting device structures,<sup>116–118</sup> and processing<sup>119–121</sup> and other techniques.<sup>122,123</sup> The silicon-based solar cells have high  $\eta$  above 20%,<sup>92,95,96</sup> but they have limitations of brittleness and difficult handling. Otherwise, other solar cells based on the coating process can be fabricated onto various electrodes, made of metal oxides (*e.g.* indium tin oxide (ITO), fluorine-doped tin oxide (FTO)), metals (Au, Ag, *etc.*), carbons (*e.g.* carbon nanotubes (CNTs) and graphene), coated substrates (*i.e.* glass, polymer films, metal wire, textile, *etc.*).<sup>124–126</sup> To fabricate “*All-in-one energy harvesting and storage devices*” through hybridization, the understanding and application of devices in terms of materials, electrodes, and processes utilized in fabrication are essential tasks, because the physical properties of flexibility, bendability, wiring, stacking, *etc.* are important for hybridization between devices.<sup>109,126,127</sup> It cannot be emphasized enough that the solar cells should be one of the most important future renewable energy sources in our daily lives, for powering electrical devices, housing, automobiles, *etc.*

### 3. Hybridization of energy harvesters

#### 3.1 Integration of various energy harvesters

Recently, hybrid energy harvesters for simultaneously harvesting multiple types of environmental energy have represented a new trend because of their synergetic output performances, and some hybrid cells have been accordingly demonstrated for harvesting mechanical and solar energy, mechanical and thermal energy, thermal and solar energy, and mechanical, thermal, and solar energy. For integrated energy harvesters of two or more such sources, electric circuit design should be considered, because solar cells and thermoelectric generators generate direct current (DC) electricity, while piezoelectric, triboelectric, and pyroelectric generators usually generate alternating current (AC) electricity (Fig. 2). In the case of AC electricity, a rectification diode is required to convert AC into DC electricity. Moreover, impedance matching depending on materials and operation frequencies using resistive load to the piezoelectric generators,<sup>128,129</sup> pyroelectric generators<sup>130,131</sup> and triboelectric generators<sup>132,133</sup> is important to achieve maximum power output (Fig. 3).

**3.1.1 Mechanical and solar energy.** Multi-type energy harvesters using both mechanical and solar energy have been especially intensively developed after a hybrid cell was first

proposed in 2009 by Wang's group, based on piezoelectric and PV effects.<sup>134</sup> The piezoelectric effect is driven by ultrasonic waves with a fixed frequency of  $\sim 41$  kHz. The range of various frequencies of mechanical energy in our living environment should be considered, however, for the study of PENGs for practical applications. Lee *et al.*, in 2010, demonstrated a hybrid energy harvester fabricated by infiltrating CdS/CdTe quantum dots into vertically aligned ZnO nanowires driven by sound in the frequency range of 35–1000 Hz and solar energy as shown in Fig. 4a and b.<sup>135</sup> After this architecture was proposed, various hybrid energy harvesters were suggested and demonstrated using acoustic waves and pressure for the piezoelectric effect, which were based on PVDF<sup>136,137</sup> and ZnO,<sup>138</sup> with various kinds of solar cells such as DSSCs,<sup>138</sup> quantum dot solar cells,<sup>135</sup> silicon (Si) solar cells,<sup>136</sup> and organic–inorganic hybrid solar cells.<sup>137</sup> Since the invention of TENGs by Fan *et al.* in 2012,<sup>9</sup> TENG-based multi-type energy harvesters have also received great attention because of their high output power and easy hybridization with other energy harvesters.<sup>139–144</sup> In 2013, Yang *et al.* demonstrated a hybrid energy harvester based on the triboelectric effect and the PV effect for the first time. Micro-pyramid Si solar cells were fabricated with a protective layer consisting of a thin film of polydimethylsiloxane (PDMS) nanowires, which not only worked as a protective layer, but also as a triboelectric layer for harvesting mechanical energy (Fig. 4c and d).<sup>139</sup> The hybrid energy harvester can be used for self-powered electrodegradation of rhodamine B and can also charge lithium (Li) ion batteries for operating small electronic devices. A Si solar cell covered with the polymer showed decreased conversion efficiency from 16% to 14%, however. In 2014, Guo *et al.* improved the structural design and device performance with airflow-induced vibration of the polytetrafluoroethylene (PTFE) film between the copper (Cu) electrodes

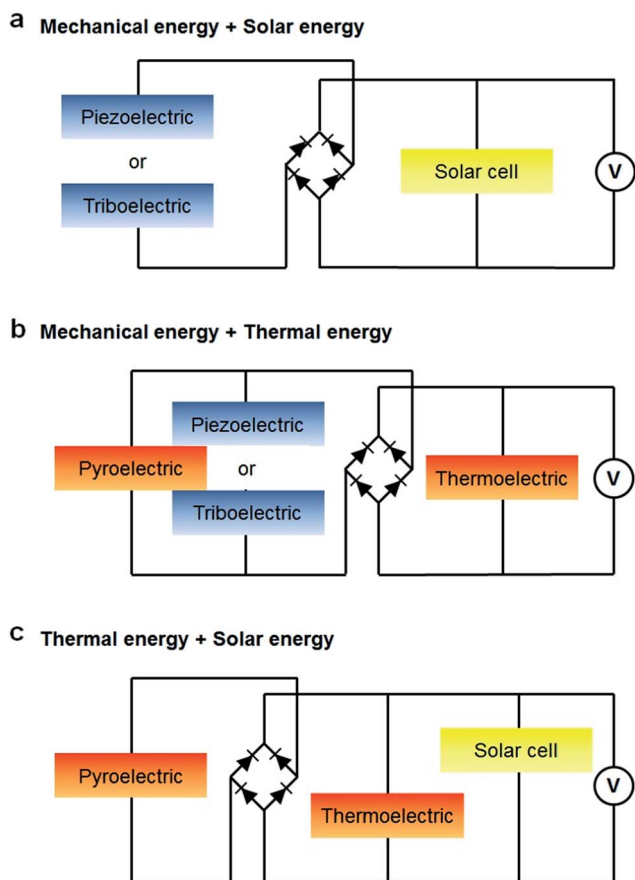


Fig. 2 Electric circuit designs for hybrid energy harvesters. (a) Mechanical energy + solar energy, (b) mechanical energy + thermal energy, and (c) thermal energy + solar energy.

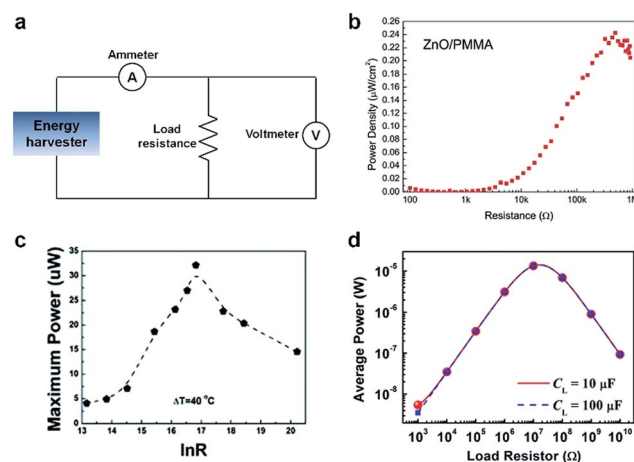


Fig. 3 (a) Typical schematic of a measuring circuit for current and voltage as a function of load resistance. (b–d) Output power as a function of load resistance: (b) ZnO based piezoelectric energy harvester, (c) PVDF based pyroelectric energy harvester, and (d) triboelectric energy harvester. (b) Reproduced from ref. 129 with permission from the Institute of Electrical and Electronics Engineers, (c) from ref. 131 with permission from the Royal Society of Chemistry and (d) from ref. 132 with permission from the Elsevier.



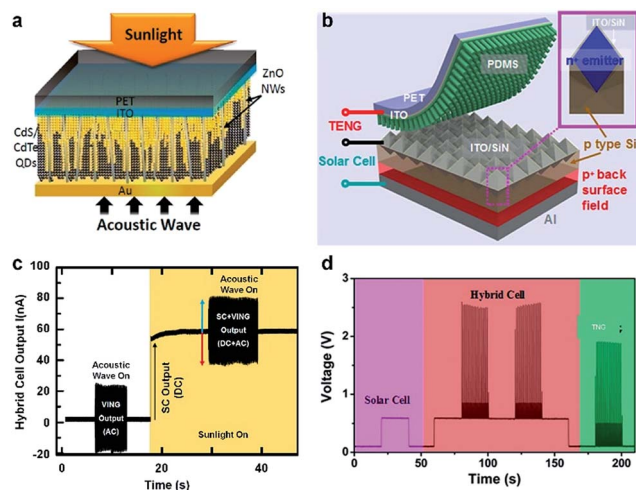


Fig. 4 (a) Schematic diagram depicting a hybrid device with two different incoming energy sources, and (b) short-circuit current output signal of the hybrid cell. (c) Schematic diagram of the fabricated hybrid energy cell and scanning electron microscope (SEM) image of the fabricated Si pyramids, and (d) output voltage of the hybrid solar cell and TENG (after rectification) for harvesting both solar and mechanical energy. (a and b) Reproduced from ref. 135 with permission from the American Chemical Society and (c and d) from ref. 139 with permission from the American Chemical Society.

and the DSSC (Fig. 5a and b).<sup>141</sup> This airflow induced TENG can effectively prevent the mechanical damage caused by direct friction between the electrodes, and output power is also much enhanced. This device is also able to drive 46 commercial green light-emitting diodes connected in series and charge a capacitor (220 mF) to 2.5 V in 50 s. Based on these demonstrations of hybrid energy harvesters for concurrently harvesting solar and mechanical energy, in 2015, Fang *et al.* reported a solution processed flexible hybrid energy harvester based on integration

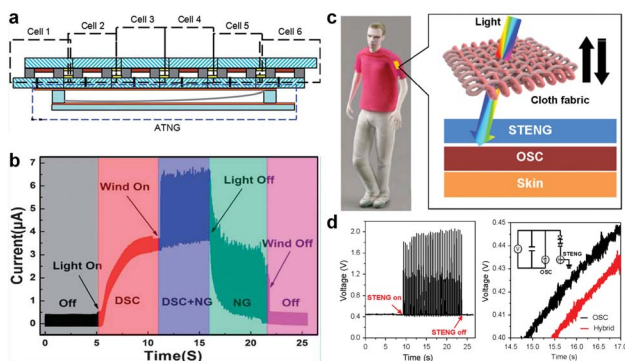


Fig. 5 (a) Schematic diagram of a hybrid device, and (b) the time-current signals of the hybrid device, with the modules working individually and simultaneously to harvest wind and light energy. (c) Schematic illustration of the working mechanism of the hybrid cell hidden under the cloth fabric as a wearable power source. (d) Time required to charge a 10 mF commercial capacitor using the hybrid cell with and without single electrode triboelectric nanogenerator (STENG) input. (a and b) Reproduced from ref. 141 with permission from the Royal Society of Chemistry and (c and d) from ref. 143 with permission from the Elsevier.

of an organic solar cell and a single electrode TENG, for use as a wearable energy harvester (Fig. 5c and d).<sup>143</sup> This prototype represents a possible approach to a power source that would work even on a rainy day or upgrade various wearable electronics, such as for epidermal healthcare monitoring without an external power supply.

**3.1.2 Mechanical and thermal energy.** In some situations, mechanical vibrations or friction, and temperature fluctuations or a temperature gradient coexist, such as on the human body, during air flow, and in working engines/machines.<sup>32,137,140,145,146</sup>

In 2014 Lee *et al.* demonstrated a stretchable piezoelectric-pyroelectric hybrid energy harvester based on a micro-patterned ferroelectric polymer P(VDF-TrFE) thin film, a PDMS-CNT composite, and graphene electrodes, as shown in Fig. 6a.<sup>32</sup> Harvesting mechanical and thermal energy from a single cell was successfully demonstrated, and the output electrical signals were integrated under simultaneous application of mechanical and thermal energy (Fig. 6b). The working mechanism for achieving the total output voltage was based on coupling of the piezoelectric and pyroelectric effects in terms of polarization and oscillation of the electric dipoles of P(VDF-TrFE). Stable pyroelectric output performance of the hybrid energy harvester for various stretchable modes was also demonstrated, which showed the stretchability, mechanical durability, and robustness of the device owing to the micro-patterned design. A Seebeck effect based hybrid energy harvester to harvest both mechanical and thermal energy was also developed to convert wasted temperature gradient energy into electrical energy. In 2013, Lee *et al.* demonstrated a hybrid energy harvester based on the piezoelectric and thermoelectric effects, by integration of the piezoelectric and thermoelectric component devices, which were mounted on a flexible substrate, as shown in Fig. 6c.<sup>145</sup> The output performance of the two processes can be integrated without loss of the combined output, including the high output current from the TEG ( $I = 5 \mu\text{A}$  and  $V = 0.45 \text{ mV}$  at  $\Delta T = 3 \text{ }^\circ\text{C}$ ) and the high output voltage from the PENG ( $I = 200 \text{ nA}$ ,  $V = 3 \text{ V}$ ), as shown in Fig. 6d and e. Furthermore, they demonstrated that the energy harvesting device can simultaneously harvest using both thermal and mechanical energy from the human body temperature gradient and mechanical movement. In 2013, Yang *et al.* also fabricated a hybrid generator that consisted of a TENG and a TEG, which could be used for self-powered water splitting to generate hydrogen (Fig. 7a).<sup>140</sup> The hybrid energy harvester consisted of a polyamide (PA)-perfluoroalkoxy (PFA) polymer film-based TENG and a  $\text{Bi}_2\text{Te}_3$ -based TEG. The fabricated TENG generated output voltage and current of 110 V and 60  $\mu\text{A}$ , and the TEG generated output voltage and current were 2.2 V and 0.1 A, respectively (Fig. 7b and c). This strategy provides a highly promising platform for harvesting both mechanical and thermal energy simultaneously and individually, and utilizing it for wireless sensors, temperature imaging, medical diagnostics, power sources for biomedical applications, personal electronics, sensor networks and micro/nano-systems.

**3.1.3 Thermal and solar energies.** Hybrid cells for harvesting thermal and solar energy are also the most conventional energy harvesting technology which has been reported.<sup>87,137,140,147-154</sup> A thermoelectric-solar hybrid generator was

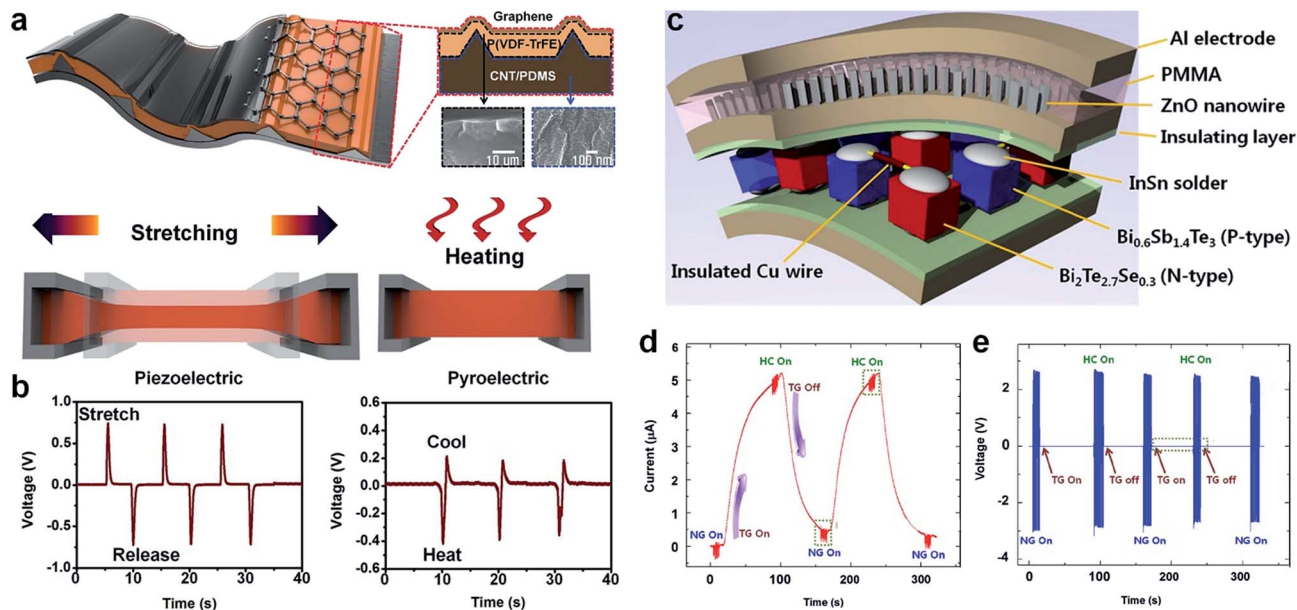


Fig. 6 (a) Schematic illustration of a hybrid device for harvesting mechanical and thermal energy; (b) piezoelectric output voltage from the device under stretch-release conditions (left), and pyroelectric output voltage under a thermal gradient (right). (c) Flexible hybrid cell, consisting of a piezoelectric generator above a thermoelectric generator, (d) output current when the TEG and the PENG are connected in parallel (left), and (e) output voltage when the TEG and the PENG are connected in series (right). (a and b) Reproduced from ref. 32 with permission from WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim and (c–e) from ref. 145 with permission from the Elsevier.

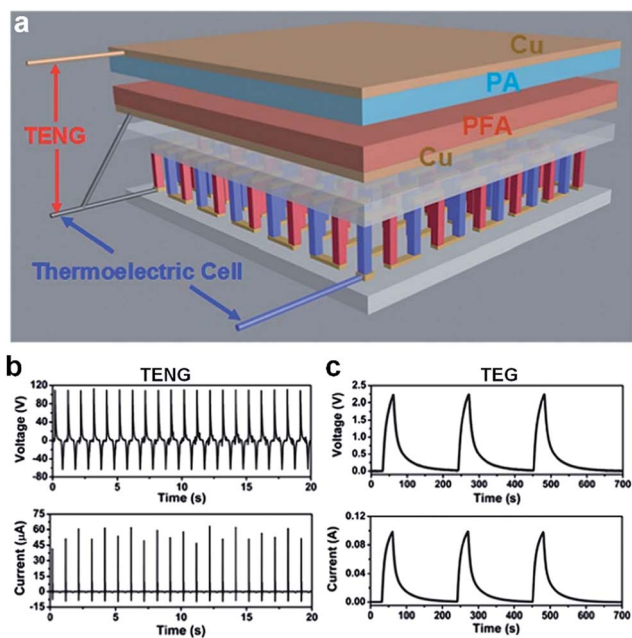


Fig. 7 (a) Schematic diagram of the fabricated hybrid energy cell consisting of a triboelectric generator and a thermoelectric cell, (b) output voltage and current of the triboelectric generator, and (c) output voltage and current of the thermoelectric generator. Reproduced from ref. 140 with permission from the Royal Society of Chemistry.

proposed in 2005 by Zhang *et al.*, using a Si solar cell and a TEG.<sup>147</sup> Sunlight collected by a lens is separated into two energy ranges of light, with the ultraviolet (UV)-visible light

absorbed by a solar cell and the infrared light absorbed by a TEG to harvest electricity. In 2010, Guo and Chang designed a thermoelectric-solar energy harvesting system to increase output performance with a DSSC and a TEG.<sup>148,149</sup> After this study, in 2011, Wang *et al.* reported a novel approach to a thermoelectric solar hybrid energy harvesting system with a series connected DSSC, a solar selective absorber (SSA), and a TEG, as shown in Fig. 8a.<sup>151</sup> The conversion efficiency of the solar cell was greatly increased by using the SSA and the TEG to utilize residual sunlight transmitted through the DSSC. The hybrid device, comprising a DSSC for high-energy photons and an SSA-coated thermoelectric generator for low-energy photons, yielded overall conversion efficiency higher than 13% (Fig. 8b). In 2013, Yang *et al.* demonstrated a hybrid cell that was composed of a  $\text{Bi}_2\text{Te}_3$  based TEG and a Si solar cell, which could be used to simultaneously and individually harvest thermal and solar energy (Fig. 8c).<sup>140</sup> Under light illumination, the output voltage of the hybrid energy cell is about 3.5 V, and the output current is about 30 mA, based on the integration of 6 solar cells (Fig. 8d). Under light illumination on the solar cells and heat applied at the bottom of the thermoelectric cell, the total peak output voltage and current of these two components of the energy harvester reached 5.2 V and 34 mA, respectively. Using the hybrid energy harvester, the Li-ion battery could be easily charged from 1 V to 3.7 V in only about 376 s. Under a constant discharging current of 5 mA, the charged Li-ion battery can continue to operate for about 327 s before it gets back to the original value of 1 V, corresponding to total electric capacity of 0.45 mA h. In 2015, Park *et al.* reported a hybrid cell based on a DSSC solar cell with a pyroelectric and thermoelectric device operated by photothermally generated heat, as shown in

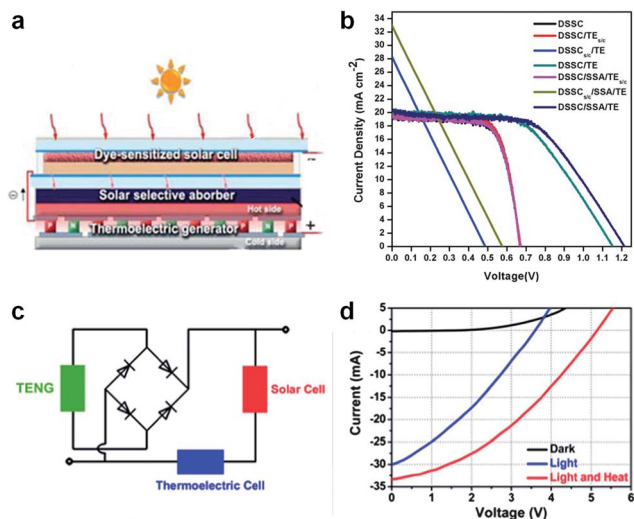


Fig. 8 (a) Schematic illustration of a solar-thermoelectric hybrid device and (b) photocurrent density–voltage ( $J$ – $V$ ) characteristic curves of the DSSC/TEG hybrid device and of the component devices with the other component short-circuited. (c) Schematic diagram of the hybrid energy cell and (d) current–voltage characteristics of the hybrid energy cell under different conditions. Reproduced from ref. 151 with permission from the Royal Society of Chemistry and (c and d) from ref. 140 with permission from the Royal Society of Chemistry.

Fig. 9a.<sup>87</sup> The photoconversion efficiency (PCE) was increased up to 20% under sunlight irradiation (AM 1.5G) using the transmitted light through the DSSC as a heat source that was converted into electricity by the pyroelectric and thermoelectric effects simultaneously by the photothermal poly(3,4-ethylenedioxythiophene) (PEDOT) electrodes. Interestingly, as PV works under sunlight, the output voltage from the PV device

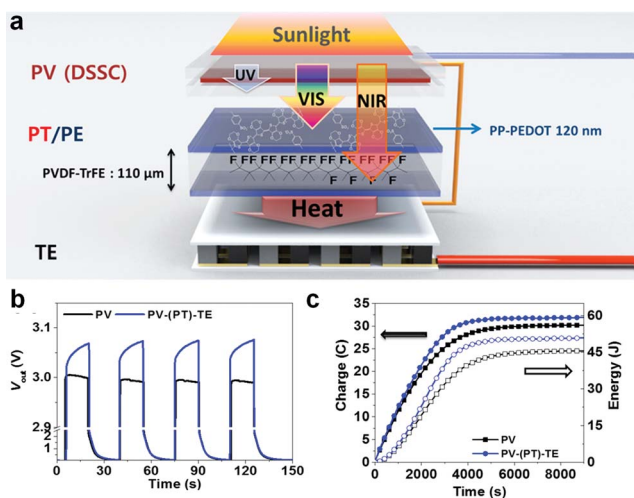


Fig. 9 (a) Illustration of a photovoltaic and photothermal pyro-thermoelectric device. (b) Output voltage of a PV and PV–TE series connected device. (c) Output and accumulated energy of a capacitor (10 F, 5.4 V) charged by PV and by the PV–TE series connected device in the hybrid system (filled circles and squares, charge; open circles and squares, energy). Reproduced from ref. 87 with permission from the American Chemical Society.

continuously decreased because of the fast recombination phenomenon during the PV operation (Fig. 9b). When the PV was combined with a TEG, however, the total output voltage increased, due to thermoelectric power generation. Consequently, the reduced PV output performance was enhanced by the output of the thermoelectric generator in the hybrid device. This result was also confirmed by the charging of the capacitor. The plot (Fig. 9c) of the cumulative charges and energies in the capacitors demonstrates the enhanced performance of the PV–TE hybrid system.

### 3.2 Coupling of multiple energy sources

Since a hybrid generator was demonstrated in 2009 by Xu *et al.*, the new concept of a piezoelectric potential enhanced PV cell was also suggested, with the mechanism called the piezo-phototronic effect. In 2009 and 2011, Xu *et al.* demonstrated that in a serially integrated DSSC solar cell and ZnO PENG, the open circuit voltage in AM 1.5G and ultrasonic waves could be enhanced.<sup>134,155</sup> The working principle is explained using the electron energy band diagram, as shown in Fig. 10. The maximum achievable output voltage is the difference between the Fermi level of the ZnO nanowires (NWS) in the DSSC and that of the ZnO NWS in the NG. The maximum output voltage is an integration of the output voltages of the NG and DSSC. Yang *et al.* also demonstrated the so-called piezotronic effect on the output voltage of flexible solar cells using poly(3-hexylthiophene-2,5-diyl) (P3HT)–ZnO microwire p–n heterojunctions on a flexible polystyrene (PS) substrate (Fig. 11a).<sup>156</sup> The open-circuit voltage  $V_{oc}$  of the solar cell was characterized by tuning the strain-induced polarization charges at the interface between ZnO and P3HT. Fig. 11b shows the mechanism of the piezotronic effect on solar cell performance under strain. The strain-induced piezoelectric potential is created under an externally applied strain, which modifies the energy band diagram at the interface of the p–n heterojunction, consequently modulating the performance of the device. The output power of solar cells could be enhanced by tuning the band profile under strain. In 2015, Yoon *et al.* reported a hybrid system consisting of a DC type PENG on a two-dimensional (2D) ZnO nanosheet and an organic solar cell based on P3HT/[6,6]-phenyl  $C_{61}$  butyric

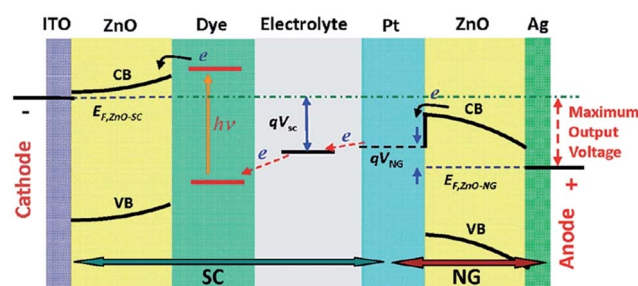


Fig. 10 Electron energy band diagram of the solar-piezo hybrid cell, showing that the maximum output voltage is the sum of the voltages produced by the solar cell and the PENG. The abbreviations are as follows: conduction band (CB), valence band (VB), Fermi level (EF). Reproduced from ref. 134 with permission from the American Chemical Society.

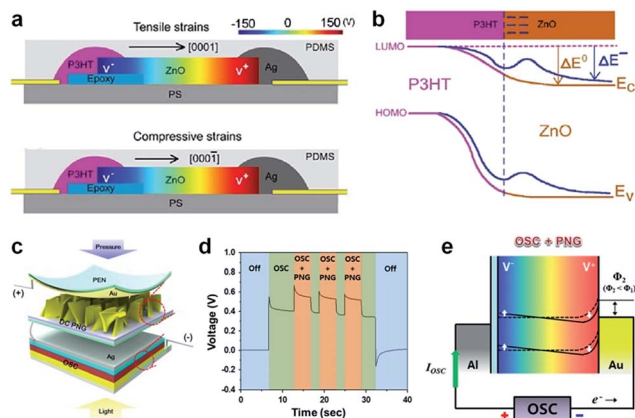


Fig. 11 (a) Piezopotential distributions in the stretched device of [0001] type and compressed device of [000 $\bar{1}$ ] type, and (b) schematic energy band diagram of P3HT/ZnO in the presence of negative piezoelectric charges. The blue line indicates the energy band diagram modified by the piezoelectric potential in ZnO. The negative piezoelectric charges can lift the energy band, resulting in a peak in the energy band. (c) Schematic illustration of the solar-piezo hybrid cell, (d) output voltage of the solar-piezo hybrid cell when pressure is applied periodically at intervals of 3.0 s for a period of 1.0 s, and (e) when pressure is applied to the PENG, the ZnO nanosheets exhibited a new band diagram: dashed and solid lines represent the band diagrams before and after applying the pressure on the PENG. (a and b) Reproduced from ref. 156 with permission from the American Chemical Society and (c–e) from ref. 157 with permission from the Elsevier.

acid methyl ester (PCBM), as shown in Fig. 11c.<sup>157</sup> The power generation performance of the serially integrated hybrid cell is synergistically enhanced by the help of a PENG, compared with the output power generated independently from the solar cell component under illumination (Fig. 11d). Fig. 11e exhibits the mechanisms of power generation from the piezoelectric potential of the ZnO nanosheets. The energy band diagram in the ZnO nanosheet is modified under the influence of applied force and light. The photogenerated electrons from the organic solar cell can effectively flow through the PENG, owing to the reduced Schottky barrier height between the ZnO nanosheets and the Au electrode. Consequentially,  $J_{sc}$  from the solar-piezo hybrid cell also increases under application of pressure.

Moreover, coupling of thermal/mechanical energy and thermal/solar energy based energy harvesters and self-powered photocurrent were also reported. In 2015, Lee *et al.* reported a PNG enhanced by piezoelectric potential driven by thermally induced strain, as shown in Fig. 12a.<sup>158</sup> Dramatic enhancement of the piezoelectric coupled PNG performance was observed, based on coupling of the piezoelectric and pyroelectric effects using different thermal expansion coefficients and micro-patterned architectures, as shown in Fig. 12b. Wang *et al.* reported using the light-self-induced pyroelectric effect in ZnO to modulate the optoelectronic processes and thus enhance the performance of ultraviolet sensors (Fig. 12c and d).<sup>159</sup> Thus, the coupling effect between mechanical, thermal, and light energy in an enhanced synergetic energy harvesting system has

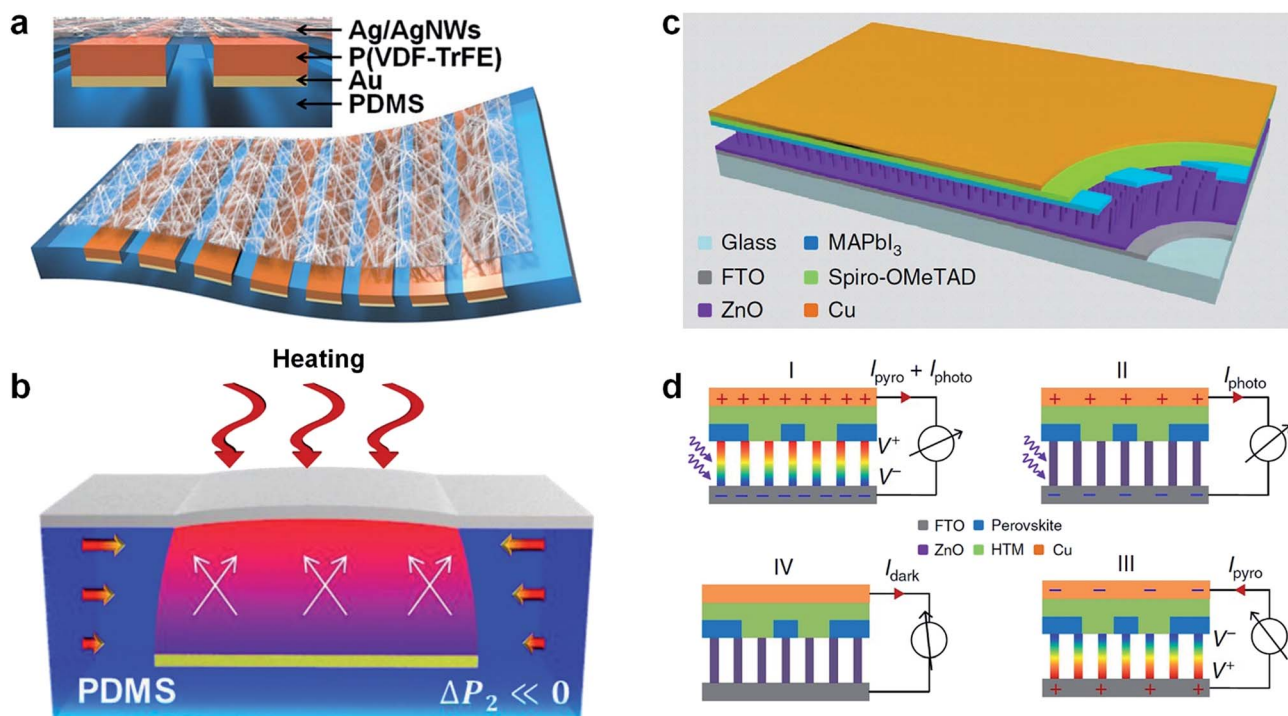


Fig. 12 (a) Schematic illustration of the piezo-pyro coupling NG, and (b) piezoelectric and pyroelectric coupled potential-generating mechanism of the patterned P(VDF-TrFE) on a PDMS substrate. (c) Schematic illustration of the structure of self-powered ZnO/perovskite-heterostructured photodetectors (ZPH PDs), with FTO acting as the transparent electrode, and (d) schematic illustration of the working mechanism of the pyroelectric effect-combined with photoexcitation processes. (a and b) Reproduced from ref. 158 with permission from WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim and (c and d) from ref. 159 with permission from the Nature Publishing Group.

attracted great attention, as it is not just simple multi-type energy harvesting, and because such devices will offer a promising approach for effectively harvesting multi-type energy for realizing multi-functional energy devices.<sup>134,138,155–164</sup>

## 4. Integration of energy harvesting and storage devices

Because of the naturally uncontrollable and unstable features of environmental mechanical, thermal, and solar energy sources, the converted electrical energy from energy harvesters is unstable and difficult to use as a direct power source for electronic devices. Usually, storage elements such as either capacitors or batteries are needed to stabilize and control the power output for direct applications.

### 4.1 Simple connection of energy harvester and storage device

The integration of energy harvesting and energy storage in one device not only enables the conversion of ambient energy into electricity, but also provides a sustainable power source for various electronic devices and systems. Therefore, there is considerable literature on the subject of various energy harvesters integrated with energy storage devices such as capacitors or batteries to demonstrate practical applications such as light emitting diodes, liquid crystal displays, electroluminescence, micro-heaters, mobile phones, sensors, pacemakers, deep brain stimulation, *etc.*<sup>165–185</sup> Solar cells and TEGs can be connected directly with energy storage devices without any electrical circuit because of their DC behaviour.<sup>186–193</sup> Nevertheless, PENGs, TENGs, and PNGs should be connected to energy storage devices with rectification diodes due to their AC behaviour. In order to effectively charge an energy storage device, the charging voltage and current should be constant with an appropriate value, whereas energy harvesters generate irregular and unstable electricity. Therefore, an optimized circuit including a capacitor filter, AC–DC and DC–DC converter is needed to economically charge the energy storage device using energy harvesters, as shown in Fig. 13.<sup>194–200</sup>

### 4.2 Hybrid structure of energy harvester and storage device

It is highly recommended that unnecessary energy loss should be minimized in the power-management circuits between

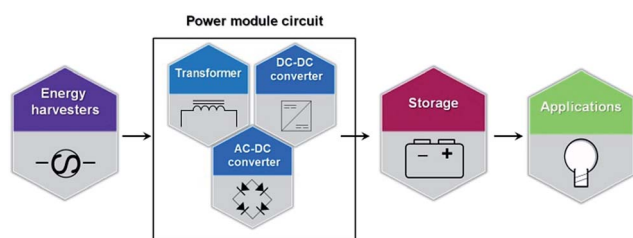


Fig. 13 Schematic diagram of an energy harvesting module circuit, which includes energy harvesters, the power module circuit, energy storage, and applications.

energy harvesting and storage devices. In general, electricity generation and energy storage are two distinct processes that involve first the conversion from mechanical, thermal, or solar energy to electricity and then the conversion from electrical energy to electrochemical energy, respectively. In 2012, Xue *et al.* first introduced a self-charging power cell using a Li-ion battery consisting of a LiCoO<sub>3</sub> cathode, TiO<sub>2</sub> nanotube anode, and PVDF piezoelectric polymer film as the separator (Fig. 14a).<sup>201</sup> The strain induced piezoelectric potential from the PVDF film acts as a charge pump to drive Li ions to migrate from the LiCoO<sub>3</sub> cathode to the TiO<sub>2</sub> nanotube anode with accompanying charging reactions at the electrodes, which can be defined as a piezo-electrochemical process. The fundamental mechanism that directly hybridizes the two processes into one, in which the mechanical energy is directly converted into electrochemical energy, without any intermediate step of first converting piezoelectric potential into electricity, is also proposed. Fig. 14b shows the charging mechanism in detail, in which the polarized PVDF film creates a positive piezoelectric potential on the cathode side and a negative piezoelectric potential on the anode side under a compressive stress on the device. Li ions in the electrolyte drift from the cathode to the anode along the pores within the PVDF film in order to screen the piezoelectric field. The decreased concentration of Li<sup>+</sup> around the cathode will break the chemical equilibrium at the cathode (LiCoO<sub>2</sub> ↔ Li<sub>1-x</sub>CoO<sub>2</sub> + xLi<sup>+</sup> + xe<sup>-</sup>), so that Li<sup>+</sup> deintercalates from LiCoO<sub>2</sub> to form Li<sub>1-x</sub>CoO<sub>2</sub>. Likewise, with increasing concentration of Li<sup>+</sup> around the anode, the chemical equilibrium at the anode (TiO<sub>2</sub> + xLi<sup>+</sup> + xe<sup>-</sup> ↔ Li<sub>x</sub>TiO<sub>2</sub>) is also broken, and the Li ions will move in the opposite direction, where Li<sup>+</sup> will react with TiO<sub>2</sub> to form Li<sub>x</sub>TiO<sub>2</sub>. During this process, Li ions migrate from the cathode to the anode continuously, and the Li-ion battery is partially charged. When the distribution of Li<sup>+</sup> can balance the piezoelectric field, a new equilibrium is achieved, and the self-

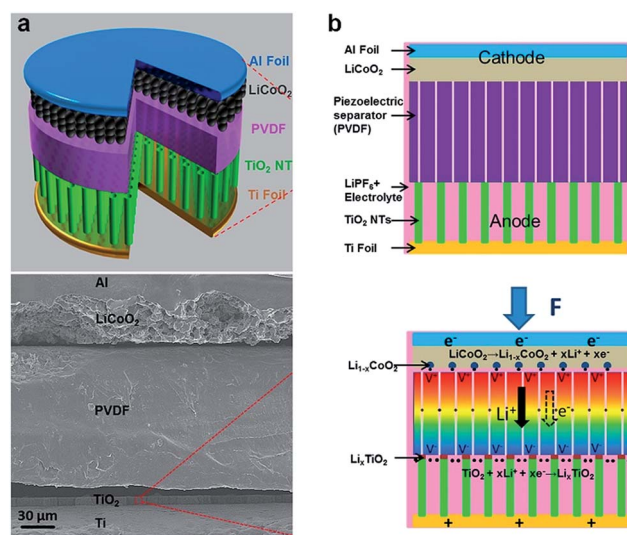


Fig. 14 (a) Design of a self-charging power cell by hybridizing a PENG and a Li-ion battery, and (b) the working mechanism of the self-charging power cell driven by compressive strain. Reproduced from ref. 201 with permission from the American Chemical Society.

charging process is completed. After this report, modification of the anode material of a lithium ion battery, using materials such as CuO,<sup>202</sup> graphene,<sup>203</sup> graphite,<sup>204</sup> and CNTs,<sup>205</sup> was demonstrated in a hybrid cell consisting of a Li-ion battery and a piezoelectric film. Zhang and Kim also reported a similar hybrid cell with the PVDF layer replaced by a PVDF-PZT nano-composite film<sup>205</sup> and a porous PVDF<sup>204</sup> membrane to enhance the charging efficiency.

In 2015, Song *et al.* and Ramadoss *et al.* fabricated a piezoelectric driven self-charging supercapacitor power cell.<sup>206,207</sup> In particular, Song *et al.* demonstrated a piezoelectric-supercapacitor hybrid cell with functionalized carbon cloth as the supercapacitor electrode, which shows the potential for wearable applications of hybrid cells, as shown in Fig. 15a.<sup>206</sup> The working mechanism of the piezoelectric-supercapacitor hybrid cell is slightly different from that of the piezoelectric-battery hybrid cell (Fig. 15b). When the external stress is applied to the piezoelectric-supercapacitor hybrid device, the remnant polarization of the PVDF film is changed. In order to balance the changed remnant polarization, charge carriers will migrate towards the electrode of the supercapacitor. Two factors are considered for the charging process. First, the increased bound charge density of the PVDF surface affects the distribution of the positive ions and negative ions in the electrolyte. The piezoelectric potential causes the redistribution of the ions and causes charges to accumulate on the surface of the supercapacitor electrode. Second, the piezoelectric potential drives the migration of hydrogen ions ( $H^+$ ) and  $SO_4^{2-}$  along the direction of the potential across the porous PVDF film. As the piezoelectric potential increases between the positive and negative electrodes due to an increasing external applied force, more electricity is stored on the electrodes in the form of electrochemical energy. The nonfaradaic and faradaic forces of the

two electrodes reach a new equilibrium after the  $H^+$  ions and  $SO_4^{2-}$  ions are redistributed in the electrolyte and balance the piezoelectric field in the PVDF film. This is an innovative approach to developing a new energy technology, which directly converts mechanical energy into electrochemical energy without energy being wasted on the outer circuitry and decreases energy conversion loss. Importantly, the development of both high performance energy storage devices<sup>208-214</sup> and the materials<sup>215-222</sup> used in the devices is essential for the fabrication of highly effective hybridized “all-in-one energy harvesting and storage devices” in the future.

## 5. Summary and perspective

In this review, we have summarized the recent progress in various types of energy harvesters and of hybrid energy harvesters, including multi-type energy harvesters, and coupling of multiple energy sources, as well as hybridization of energy harvesters and energy storage devices for self-powered systems. We believe that this review could be useful for designing energy harvesters, storage devices, and self-powered systems. To date, great advances have been achieved in developing various types of energy harvesters based on the piezoelectric, triboelectric, pyroelectric, thermoelectric, and photovoltaic effects in various energy conversion materials. The output performance of energy harvesters has reached the point where they are able to replace currently available batteries in portable electronics for self-powered systems. Hybrid cell technologies have been demonstrated that can simultaneously generate electricity everywhere and at any time, using multiple sources of environmental energy such as mechanical, thermal, and solar energy. In particular, energy harvesters based on the coupling of multiple energy sources exhibit not only multiple energy harvesting, but also enhancement of energy harvesting performance due to synergetic effects. Finally, integration of energy harvesters and energy storage devices for practical applications such as self-powered systems was introduced. Self-charging power cells provide an especially innovative approach to hybridizing an energy harvester with an energy storage unit, which can directly convert mechanical energy into electrochemical energy without energy being wasted on the outer circuitry so as to decrease energy conversion loss. The development of technologies for the direct conversion of mechanical, thermal, and solar energy into electrochemical energy is suggested as an important topic for future work towards sustainable and maintenance-free operation of micro-/nano-systems and mobile/portable electronics. In addition all-in-one technology could be applied for the development of self-powered wearable electronics by integration of wearable energy harvester/storage devices.<sup>177,184,193,223-232</sup>

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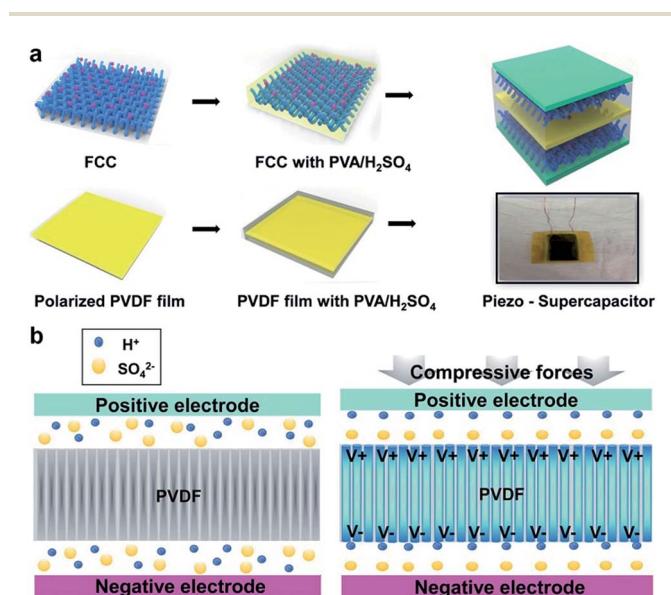


Fig. 15 (a) Schematic illustration of the process flow of hybrid piezo-supercapacitor fabrication, and (b) proposed working mechanism of the hybrid piezo-supercapacitor. Reproduced from ref. 206 with permission from the Royal Society of Chemistry.

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