Materials for Solar Cells and Supercapacitors

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Hybrid systems have gained significant attention among researchers and scientists worldwide due to their ability to integrate solar cells and supercapacitors. Subsequently, this has led to rising demands for green energy, miniaturization and mini-electronic wearable devices. These hybrid devices will lead to sustainable energy becoming viable and fossil-fuel-based sources of energy gradually being replaced. A solar photovoltaic (SPV) system is an electronic device that mainly functions to convert photon energy to electrical energy using a solar power source.

PV solar cell | supercapacitor | energy storage | integrated device | power conversion efficiency | levelized cost of energy (LCOE)

1. Introduction

The conventional supercapacitor-charging method using photovoltaic (PV) was originally designed using a solar cell and supercapacitor to operate as two independent units that are connected by wires. Despite being able to simultaneously generate and store energy, the system faces some technical challenges, such as being bulky, inflexible, expensive and, in particular, it loses energy through the external wires connecting the two units [1]. Such disadvantages can be overcome by integrating the supercapacitor and PV cell into one device so that it is compact, flexible, modular and minimizes energy loss, as no wires are required to connect the two units. This invention is in line with the current smart technology that allows for volume minimization, practicality and flexibility such that it can operate in various industries, such as small-scale and large-scale consumer electronics, electric vehicles, smart grids and wearable sensors [2][3][4]. Previously, batteries were used to integrate PV cells, but due to some inadequacies, such as slow charge/discharge capability and short life cycles, supercapacitors are preferred for integration with PV cells. This is attributed to its rapid charging/discharging ability since, first, there is no chemical reaction required, and second, the energy density is reported to be about ten times higher than batteries with the same weight [4][5]. **Figure 1** depicts a comparison between batteries, capacitors, supercapacitors and other energy storage devices in terms of specific energy, specific power and the charge/discharge time.

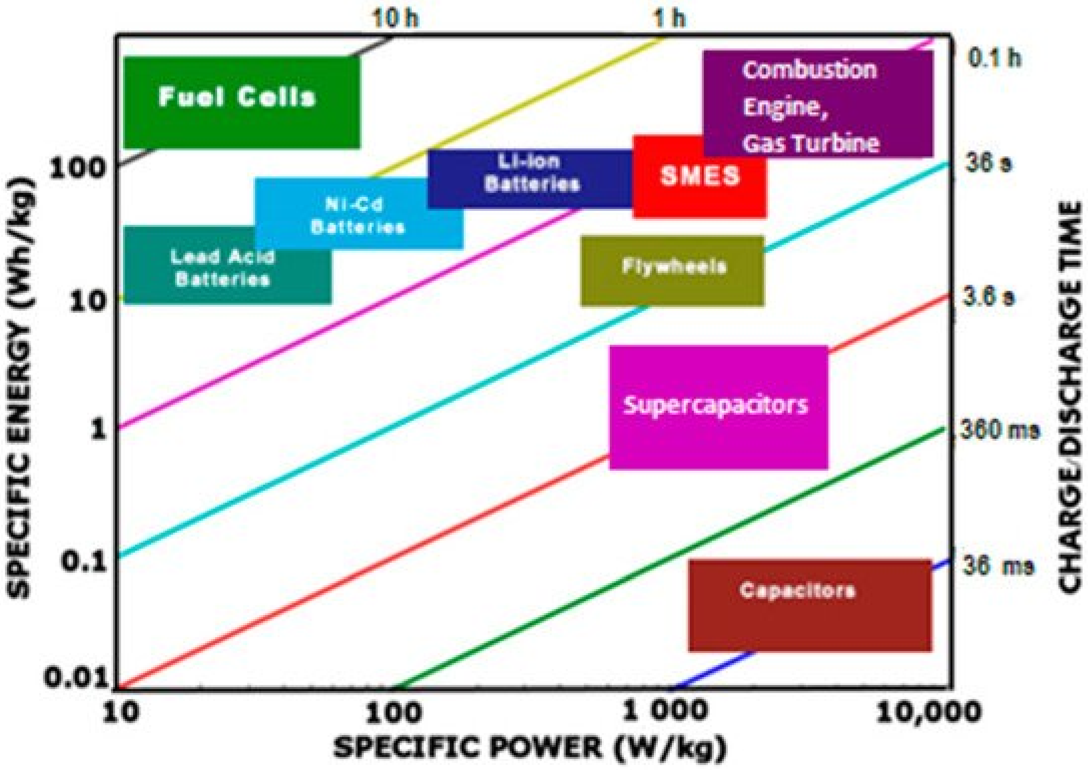

Figure 1. Ragone plot of the properties of various power sources.

Since the concept of PV cell–supercapacitor integration was introduced in the year 2004, it has sparked interest in many researchers to carry out experimental and simulation work on the fabrication of this device and assess its performance. For instance, a study by [5] integrated hydrogenated amorphous silicon (a-Si/H) solar cells, $NiCo_2O_4$ battery supercapacitor hybrids (BSHs) and f light emission diodes (LEDs) into one system. The whole system could work independently with an overall efficiency of 8.1% and the storage efficiency reached 74.24%. These values indicated that the self-driven integrated system was stable for PV conversion and had efficient energy storage. The fundamentals of supercapacitors and solar cells need to be analyzed accordingly before they are integrated into one system in order to achieve high efficiency. **Figure 2** below shows the schematic diagram of the integrated device.

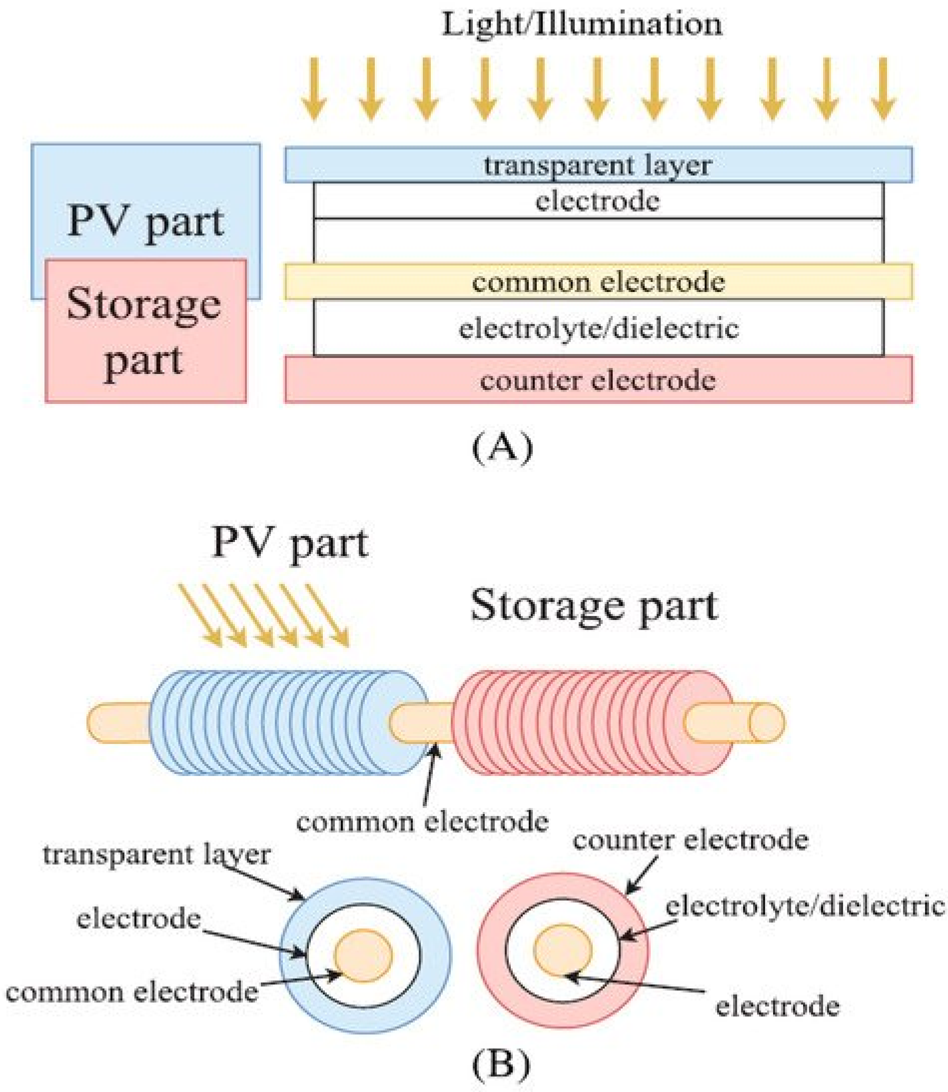

Figure 2. The schematic diagram of (**A**) planar/monolithic three-electrode system and (**B**) coaxial fiber parallel system. Reprinted from [6].

A supercapacitor's performance highly depends on the electrode materials that are used. Hence, investigation on the electrode materials for supercapacitors has been a very important topic in this research area. As technology changes, which it currently does at a rapid rate, consumers greatly rely on energy technology to cater to their electrical/electronic needs. Solar energy is a renewable energy resource and can be harnessed from all parts of the world where the relevant technology is viable. Solar energy is obtained by converting sunlight into electrical energy and it can serve industrial and domestic purposes. An energy storage system is needed to store this electrical energy in order to avoid energy loss during its operation and the supercapacitor comes in handy due to its fast charging/discharging properties alongside its long life cycle. When compared to batteries as energy storage systems, supercapacitors possess higher energy conversion with a low equivalent series resistance; these values have made supercapacitors a very suitable device for energy storage applications for solar cell panels [7].

Adding the energy storage part will increase the thickness of the cell. However, it is still more preferable than combining the two units separately because of the overall compactness and reducing the whole system's volume.

It was established that external connection was often associated with elevated resistances [8], rigid/heavy devices [9] and having a complicated manufacturing process [10]. It is worth noting that the main challenge of developing supercapacitors is increasing their energy and power densities without compromising their long life cycle and fast charging/discharging properties [10][11]. Consequently, the integration of these devices is feasible and could enhance their performance. **Table 1** below highlights the overall efficiency and storage efficiency of the PV–storage system integrated devices that were discussed in previous studies. Research on the integrated device from various materials and integration techniques reported storage efficiencies ranging from 20 to 80% and an overall efficiency of up to 8%.

Table 1. The storage and overall efficiencies of integrated devices that were documented in prior studies.

Energy Storage Device	Storage Efficiency (%)	Overall Efficiency (%)	Refs.
Ruthenium(IV) Oxide (RuO_2)//RuO_2 Supercapacitors (SCs)	26.67	0.8	[12]
Titanium@Titanium dioxide//Carbon nanotube (Ti@TiO_2//CNT) SCs	75.7	1.2	[13]
Ti@TiO_2//Multiwalled carbon nanotube (MWCNT) SCs	65.6	0.82	[14]
Carbon//carbon SCs	46.77	2.9	[15]
Poly(3,4-ethylenedioxythiophene (PEDOT)//carbon SCs	73.78	4.7	[16]
Carbon//carbon SCs	79.78	7.1	[17]
$NiCo_2O_4$//active carbon SCs	74.24	8.1	[5]

Storage efficiency is defined as the percentage of maximum energy that is used from a total energy capacity and it can be calculated based on Equation (1):

$$\text{Storage efficiency (\%)} = \frac{\text{maximum used energy}}{\text{total energy capacity}} \times 100$$

Meanwhile, the overall efficiency (η_{SS}) of an integrated device can be calculated based on Equation (2) when both efficiencies of the solar cell and energy storage device are provided:

$\eta_{SS} = \eta_{SC}\eta_{S}$(2)

where η_{SC} is the solar cell efficiency and η_{S} denotes the energy storage efficiency. Apart from that, the overall or total efficiency of the integrated device can be calculated using Equation (3):

$\eta_t = E_{output}/E_{input} = E_S S_1/(P_{input} t S_2)$(3)

where η_t is the total energy storage of the integrated device, E_{output} denotes the energy output of the supercapacitor, E_{input} refers to the total incident light energy, S_1 represents the surface area of the supercapacitor, P_{input} is the illuminated light density, t signifies the photocharging time and S_2 is the active area of the hybrid solar cell. What was observed is a huge improvement in the efficiency of the devices ranging from 10% to 80% for the storage efficiency and 1 to 11% for the overall efficiency.

It cannot be denied that smart technology is growing at a huge rate due to the rapidly changing nature and high demands of the electronics industry. Since they require great flexibility and portability of energy storage components, many researchers now want to create flexible devices that are aimed toward various applications. For instance, a high-performance hybrid supercapacitor was fabricated using 3D dendritic cell-like nanostructures with Ni-Co layered double hydroxide (LDH) used as the cathode, while the anode was a crumpled leaf-like reduced graphene oxide. The supercapacitor was integrated with a solar cell that harvested the energy and the working potential window obtained from the study was ~1.4–1.8 V. Meanwhile, the power and energy densities of the device at a current density of 0.5 A/g were ~374 W/kg and ~58.4 Wh/kg, respectively. This finding strongly suggested that high energy and power density were achievable when combining these two devices asymmetrically [18]. The supercapacitor demonstrated a superior performance since the coulombic efficiency was approximately 100% [19].

Based on the reviewed studies on this topic, it can be observed that solar cells absorb solar energy and subsequently convert it to electrical energy by using a supercapacitor as the energy transport system. Choosing appropriate active materials for the fabrication of the integrated device is crucial to maximizing the conversion efficiency. In particular, the other parameters that should be paid serious attention are the counter electrodes, conducting polymers, photoactive metal oxides and redox electrolytes given that these materials contribute to high energy conversion efficiency and would subsequently enhance the performance and shelf life of a PV cell integrated supercapacitor. Since the integrated device showed promising results in terms of its efficiencies and flexibility, this sparked great interest amongst researchers worldwide to conduct further analyses when developing solar-cell-integrated supercapacitors with improved properties.

Since improvements in the properties of PV cells and supercapacitors are widely studied and researched, integration of these two components is a novel technique to further enhance the device, particularly in terms of its conversion efficiency and storage capacity. Considering the advantages and disadvantages of PV cells and supercapacitors, these two entities are suitable for integration in order to complement the properties of the energy harvesting and storage system. The integrated device should be designed in such a way that the architecture is practically feasible as an energy generator and storage system [20].

2. Solar Cells

2.1. Materials for Solar Cells

Photovoltaic technology has evolved over the past few decades to address the challenges of converting solar energy to electricity. The main parameter to be considered for the installation of a PV cell in any device is its conversion efficiency. It is well noted that not all sunlight that reaches the PV panel will be converted into electricity. Hence, some of the parameters of a PV panel, such as the wavelength, recombination of electrons, temperature and light reflection, are crucial for the best possible conversion efficiency [21]. By considering all these parameters, the PV panel would be able to optimally convert solar energy into usable electricity for a wide range of applications. The power that is harvested from PV cells differs according to the region because the variation in solar insolation and seasons during a given year would greatly affect its performance [22]. To date, a wide variety of solar cells with different characteristics have been fabricated, namely, organic solar cells, perovskite solar cells (PSCs), dye-sensitized solar cells (DSSCs), Cu (In, Ga) Se_2 (CIGS) solar cells, etc. [7][22][23].

Recently, dye-sensitized solar cells (DSSCs) have been receiving attention, mainly due to their flexibility and cost-effectiveness. A previous study fabricated a flexible printable DSSC/supercapacitor integrated energy device that is flexible, lightweight and portable such that it can be used for a wide range of applications. It possesses a high voltage capacity that could go up to 1.8 V and is environmentally friendly. Outdoor testing was carried out with extreme mechanical loading to test its stability. The results demonstrated that the device achieved stable performance throughout the test [23]. Another researcher developed a flexible supercapacitor by doping graphene onto activated carbon. The supercapacitor that performed as an energy storage system was integrated with DSSC to power an LED. The highest capacitance was obtained by the supercapacitor doped with 0.05 wt.% graphene with a charge/discharge efficiency of 85.29%. A bending test was conducted to analyze any variation in capacitance. The results showed that the capacitance was maintained throughout the test [7].

Meanwhile, Xu and co-workers integrated an $MAPbI_3$-based perovskite solar cell (PSC) and a PPy-based supercapacitor as an energy pack [14]. The voltage of the supercapacitor was set at 0.6 V and the system produced a high output voltage of 1.45 V and an overall output efficiency of 20%. This system provided a continuous output of electric power by using the $MAPbI_3$-based solar cells as an energy source. Another PSC-integrated device was developed by [24]. A semitransparent PSC was integrated with an electrochromic WO_3 supercapacitor in a vertically stacked configuration. The power conversion efficiency that was achieved by the PV part was 8.25% and the energy density was 35.9 mW/h. The average power density and areal capacitance were 461.5 mW/m^2 and 459.6 F/m^2, respectively.

One of the established solar cell devices is made of crystalline silicon, which is still dominating the commercial PV modules in the industry despite its heavy weight, inflexibility and high production cost. Prior research by [25] integrated a commercial polycrystalline silicon device with a supercapacitor, which resulted in an 84% coulombic efficiency. The design demonstrated that the polycrystalline silicon solar cell was capable of charging the supercapacitor under an external load and that a constant current load could be maintained through periods of intermittent illumination, indicating the feasibility of the integration concept. Another type of solar cell that is widely used these days is an organic solar cell (OSC). OSCs have potentially low costs and are suitable for omnipresent distribution. Research by [26] integrated OSCs based on poly(3-hexylthiophene):[6,6]-phenyl-C60-butyric acid methyl ester with Al electrodes and supercapacitors based on graphene ink. The materials were grown on a single substrate using graphene as a common platform. The achieved power conversion efficiency (PCE) was approximately 1.6%, while the Voc was 5 V. The OSC-integrated supercapacitor managed to yield an overall system voltage of up to 4 V.

2.2. Performance of Solar Cells

There are four basic parameters that are used to evaluate the performance of solar cells, namely, the short-circuit current (I_{SC}), open-circuit voltage (V_{OC}), fill factor (FF) and PCE. The maximum power point (P_{MPP}) of a solar cell is the point where the solar cell should be operated to obtain the maximum power output, as shown in **Figure 3**.

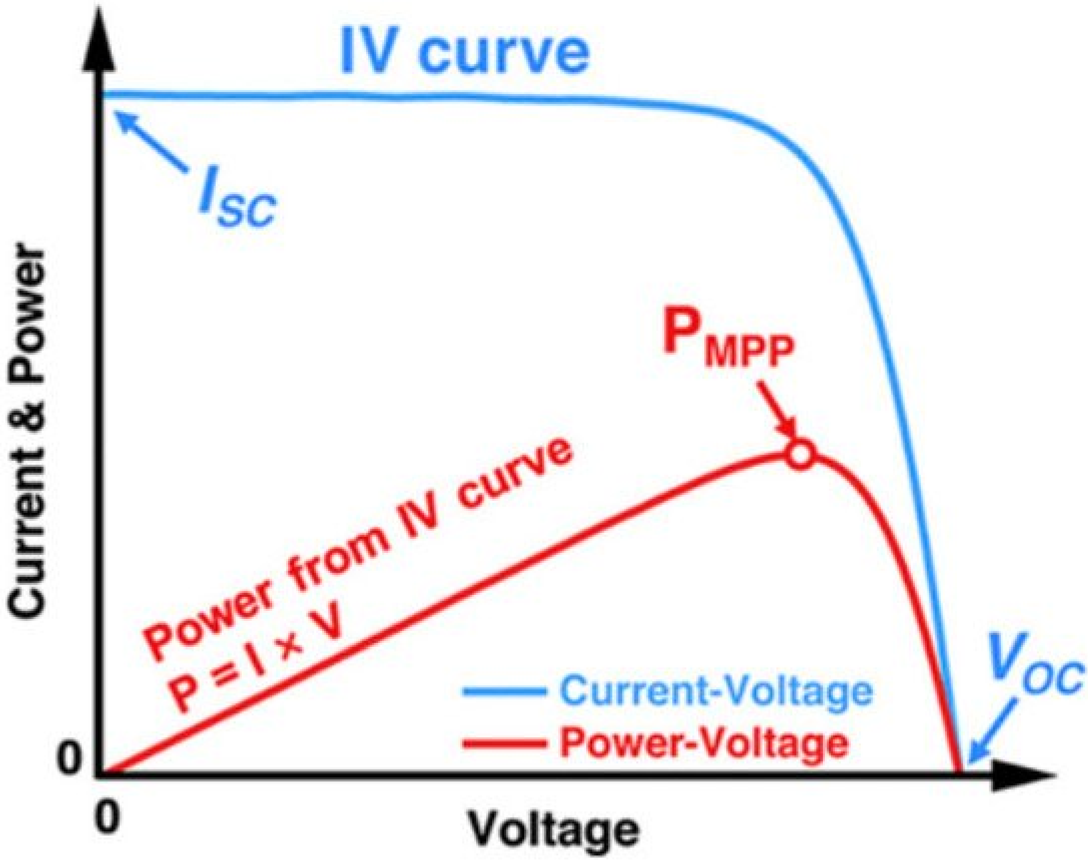

Figure 3. The I–V and power–voltage (P–V) curves of a solar cell. Reprinted with permission from reference [27]. Copyright Elsevier Publishing 2020.

It is essential to work on reducing the solar power cost; this can be done by improving the PCE of solar cells [28][29]. Data from past research showed that single-junction solar cells reached the theoretical limit of PCE and it is quite difficult to be improved further. Therefore, researchers came up with the method of stacking two or more solar cells occupying different band gaps (Eg) and optimized the wavelength absorption range. The findings showed that the theoretical PCE could surpass 50% depending on the number of cells that were stacked [30]. Previous work reported the PCE value to be approximately 40% by developing III-V multi-junction cells. However, the limitation for the use of III-V semiconductor-based tandem devices is due to its high production and material costs, which makes it applicable to only spacecraft and satellites at present [31]. The PCE is the most important parameter for solar cells, other than its efficient preparation technologies and flexible structures. **Table 2** summarizes the power conversion efficiency of the various types of solar cells from previous studies.

Table 2. Power conversion efficiencies of various types of solar cells.

Material	Power Conversion Efficiency (%)	Reference
Perovskite/Cu (In, Ga) Se_2 (CIGS)	28	[32]
Organic–inorganic perovskite formamidinium tin iodide	19.08	[33][34]
Ternary polymer solar cell	15.5	[35]
Triboelectric nanogenerator/silicon (TENG/Si) tandem hybrid solar cell	22.4	[36]
Gallium arsenide solar cell	30.6	[37]
Silicon heterojunction solar cells	26	[27]
Piezo-phototronic multijunction solar cells	33	[38]
Lead iodide perovskite-based solar cells	25	[39]

The fabrication of various types of solar cells and the combination of materials showed a significant improvement in their power conversion efficiencies as the technology is advancing and demand from industries keeps increasing. This simultaneously allows researchers to come out with notable techniques and technology to fill the gap for each problem that is faced when building solar cells. To date, substantial growth has been achieved in the development of various types of flexible solar cells, where researchers managed to increase their conversion efficiencies due to technological advancements that are crucial for the development of solar cell integrated devices.

3. Supercapacitor as the Energy Storage Component for an Integrated Device

Supercapacitors are mainly classified into three types, which are electric double-layer capacitors (EDLCs), pseudocapacitors and hybrid capacitors. EDLCs operate by storing the charge at the surface electrode through reversible ion absorption/desorption to form an electrical double-layer capacitance [40]. It was noted that an EDLC can be used as a substitute for rechargeable batteries owing to its capability of fast charging/discharging, especially for a device that necessitates rapid energy harvesting. A pseudocapacitor, on the other hand, operates using fast and reversible redox reactions on or near the electrode surface. The charge passes across the double layer and results in a faradaic current passing through the supercapacitor cell, which is termed pseudocapacitance [41]. Meanwhile, a hybrid capacitor is a combination of a porous carbon electrode and another material, such as a conducting polymer, metal oxides or metal-doped carbons, which are used to enhance the performance of EDLCs and pseudocapacitors [42]. **Table 3** summarizes the performance evaluation of different types of supercapacitors. The main challenge regarding supercapacitors is to increase the energy densities while maintaining their long life cycle, high power density and fast charging/discharging. The commercialization of energy storage devices based on hybrid supercapacitors has been a viable option to manufacturers, which highlights the importance of these new materials for a wide range of applications.

Table 3. Performance parameters for different types of supercapacitors.

Types	Operating Voltage (V)	Energy Density (Wh/kg)	Power Density (W/kg)	Refs.
Electric double-layer capacitor	Up to 3.5	~3–5	~900–10,000	[40]
Pseudocapacitor	Up to 2.3	~1–10	~500–5000	[43]
Hybrid capacitor	~3.8–19	~8–80	~200–1500	[44]

Materials for Supercapacitor Electrodes

Carbon materials, such as carbon nanotubes (CNT), graphenes and MXenes, have been widely utilized as supercapacitor electrodes due to their high specific area and outstanding conductivity. Generally, CNTs possess a tensile strength of 100–200 GPa [45], electrical conductivity of ~10^7 S/m and thermal conductivity of ∼2000 W/m·K [46]. A past study by [47] fabricated an all-solid-state integrated device from free-standing and aligned carbon nanotube films. The integrated device exhibited an overall photoelectric conversion and storage efficiency of approximately 5.12% owing to its aligned structure and outstanding electronic property of the film electrode. In addition, the device's flexibility makes it suitable for a wide range of applications, especially in portable electronic equipment. Another promising finding was obtained by [48] after integrating a DSSC with a supercapacitor by using a CNT film as the common electrode. The authors reported a PCE of 6.1%, a specific capacitance of 48 F/g and a storage efficiency of about 84%. The device is lightweight, flexible and the overall photoelectric conversion and storage efficiencies that were achieved were about 5.12%. Graphene, on the other hand, is synthesized using various methods, such as mechanical exfoliation, chemical exfoliation [49], chemical vapor deposition [50], chemical synthesis [51] and microwave synthesis [52]. Due to its high specific surface area, graphene is found to be more efficient at storing electrostatic charges, thus making it suitable for use in supercapacitor electrodes [53][54]. Even though graphene is considered a new and emerging material, extensive research has been carried out to incorporate graphene into the solar cell–energy storage integrated system. Recently, a DSSC employing a compact and mesoporous titania (TiO_2) film as the anode was integrated with a symmetrical supercapacitor utilizing polypyrrole/reduced graphene oxide (PPy/rGO) electrodes as the counter electrode. The specific capacitance of the supercapacitor was 308.1 F/g and the PCE of the DSSC was reported to be 2.4%. The integrated device exhibited a specific capacitance of 124.7 F/g, and a retention percentage of 70.9% was obtained after 50 consecutive charge/discharge cycles [55]. Meanwhile, a new emerging material, namely, MXene, has been extensively studied owing to its tremendous potential for supercapacitor electrodes [56][57]. For instance, prior work by [58] integrated a flexible organic photovoltaic with Ti_3C_2Tx MXene as the electrode and organic ionogel as the electrolyte. The authors obtained a high power conversion efficiency of 13.6% and a high volumetric capacitance of 502 F/cm^3. This simple fabrication method achieved a remarkable storage efficiency of 88%. Another related study developed wearable electronics by integrating an MXene/black phosphorus-based supercapacitor with a solar cell. The detailed schematic diagram of the integrated system is depicted in **Figure 4**. The developed MXene/BP-based micro-supercapacitor showed a high specific capacitance of 896.87 F/cm^3 and an excellent rate performance of 241.2 F/cm^3. The system also had long-term cycling stability of 91.74% for 10,000 cycles.

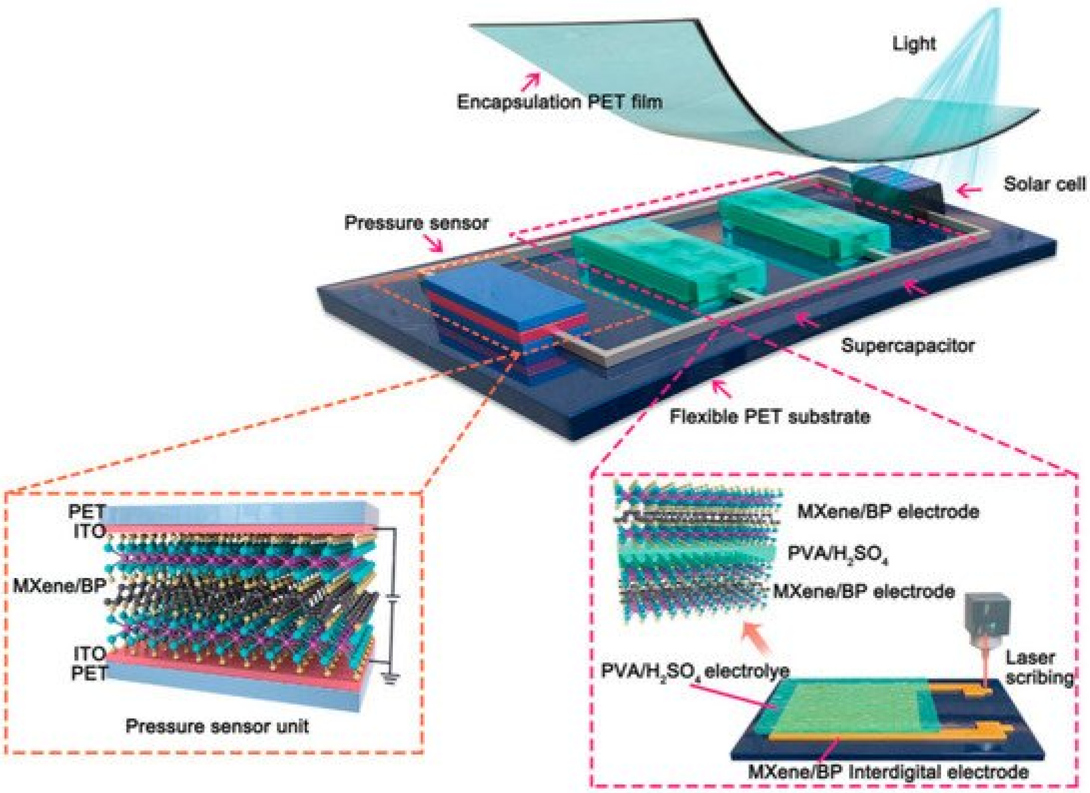

Figure 4. Schematic illustration of MXene/black phosphorus-based self-powered smart sensor system. Reprinted from [59].

The data on studies of traditional materials for supercapacitor electrode materials are widely reported and researchers have come out with more studies on new emerging materials. A summary of conventional and new materials is listed in **Table 4**.

Table 4. Traditional and emerging new electrode materials for supercapacitors [47][60][61][62][63][64][65].

Electrode Material	Examples	Properties
Nanocarbons	CNT, Activated Carbon (AC), Graphene	Chemical stability, high exohedral surface area, high electrical conductivity due to covalent sp^2 bonds, high cost
Conducting polymers	Polypyrrole (Ppy), Polyaniline (PANI), Poly(3,4-ethylenedioxythiophene	More versatile, outstanding specific energies

Electrode Material	Examples	Properties
	(PEDOT)	
Metal oxides	MnO_2, Nb_2O_5, V_2O_5	High theoretical capacitance, rapid faraday redox reaction, high cost, toxicity concern
Metal nitrides	Vanadium Nitride (VN), TiN, Fe_2N	Outstanding electrochemical properties, high chemical stability, standard technological approach
MXenes	$Ti_3C_2T_x$	High number of active groups, large surface area, high chemical reactivity
Metal-organic frameworks	Cu-catecholate (Cu-CAT)	Large surface area, three-dimensional porous architecture, permeability to foreign entities, structural tailorability
Black phosphorus		Large theoretical capacity, high carrier mobility, low redox potential
Polyoxometalates	PMo_{12}, PV_2Mo_{10}	High stability of redox states, able to participate in fast reversible multielectron transfer reactions

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