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Review

Banana Peel and Conductive Polymers-Based Flexible Supercapacitors for Energy Harvesting and Storage

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Abstract: Flexible supercapacitors are highly demanding due to their wearability, washability, lightweight property and rollability. In this paper, a comprehensive review on flexible supercapacitors based on conductive polymers such as polypyrrole (PPy), polyaniline (PANI) and poly(3,4-ethylenedioxtthiophne)-polystyrene sulfonate (PEDOT:PSS). Methods of enhancing the conductivity of PEDOT:PSS polymer using various composites and chemical solutions have been reviewed in detail. Furthermore, supercapacitors based on carbonized banana peels and methods of activation have been discussed in point. This review covers the up-to-date progress achieved in conductive polymer-based materials for supercapacitor electrodes. The effect of various composites with PEDOT:PSS have been discussed. The review result indicated that flexible, stretchable, lightweight, washable, and disposable wearable electronics based on banana peel and conductive polymers are highly demanding.

Keywords: supercapacitor; electrical conductivity; energy storage; banana peel; e-textiles

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1. Introduction

The energy sector is one of the substantial conducer crossways in the sphere for the resiliency and easy maintenances of the human being. The use of fossil fuels brings deterioration to the environment [1]. However, a study reviled that in this scenario, fossil fuels are the leading energy sources at least until 2025 [2]. The study conducted in Ref. [3] indicated that industries need high demand of energy in the future. Therefore, to utilize sustainable energy sources, researchers must develop new means of energy sources which have flexible, lightweight, and durable features.

The rapid upsurge in the world's energy usage and the impact outdated energy resources to the surrounding environment have led to the enormously augmented investigations at activities on renewable and clean energy resources during the last many decades. Therefore, there is a persistent necessity to develop unconventional energy harvesting and storage systems. Among these, supercapacitors are the most researched items as efficient energy storage devices.

With the advent of new polymers, state-of-the-art processing technologies, and renewable and sustainable methods, the growth of using flexible and sustainable energy storage materials by producing supercapacitors has increased in recent years [4,5]. Supercapacitors can be defined as materials capable of energy storing and conversion which supposed to have the potential of high-power density, great circulation feature, quick discharge-charge, and poor self-discharging characteristics [6]. Supercapacitors can be produced using several principles and methods such as using activated carbons [7,8] conductive polymers [9,10],

graphene [11], carbon nanotubes (CNT) [12], and others. Rollable, flexible, and bendable microsupercapacitors (MSCs) can be produced by mixing with phosphoric acid with SWCNT [13]. The MSCs produced found to be potential applications in wearable devices. When bending, the electrical conductivity of the substrate interrupted and hence the supercapactive performance reduced. Beyond chemical activation, adding heteroatoms also affects the performance of supercapacitors [14]. The study indicated that there are lots of possibilities to produce carbon-based supercapacitors by doping with various heteroatoms such as fluorine, phosphorous, boron, chlorine, and silicon.

Supercapacitors are specially designed capacitors which have huge capacitance value and energy density when compared to the conventional capacitors that are with fast storage ability and high energy density than capacitors [15]. Based on the working and energy storage principle, supercapacitors are categorized into three basic groups as shown in Figure 1. Electric double-layer capacitors (EDLCs) are types of capacitors constructed using three materials the so-called electrodes, electrolytes, and a separator. They are portable, very efficient, and high-power energy storage devices [16]. EDLCs store energy by means of non-faradic principle or electrostatically that encompasses no transmission of charge between the electrolyte and electrode.

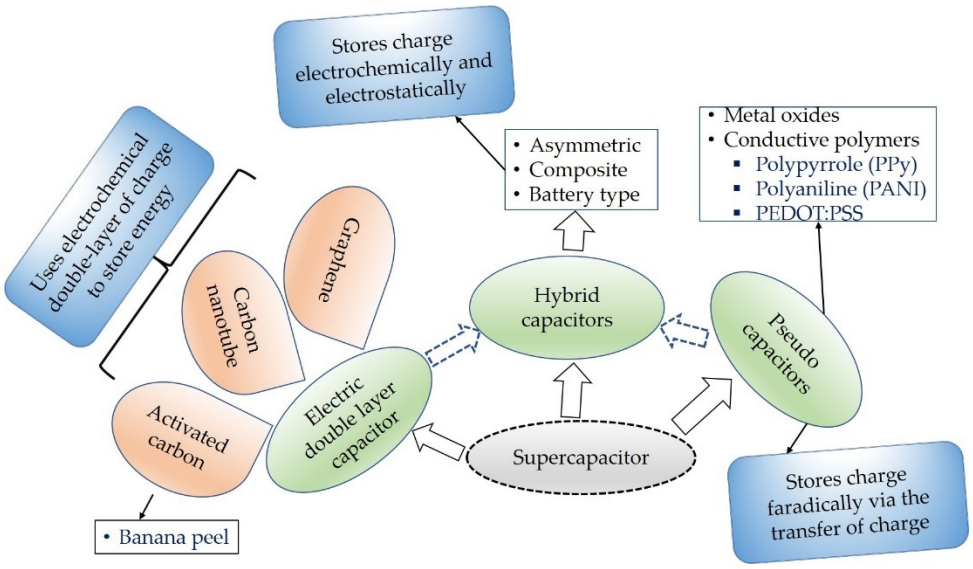

Figure 1. Types of supercapacitors based on its working principles.

Pseudocapacitors on the contrary to EDLCs store charge via the transfer of charge amongst the electrode and electrolyte faradically [17]. Pseudocapacitors employed various conductive polymers such as polypyrrole (PPy) and polyaniline (PANI) [17], poly(3,4-ethylenedioxtthiophne) polystyrene sulfonate (PEDOT:PSS) [18]. Due to the charging and discharging behavior and the reduction-oxidation rection occurred during processing, pseudocapacitors lack stability. Stability against the environment is very crucial for such types of materials.

Hybrid supercapacitors for energy storage principle is based on the combinations of the EDLCs and pseudocapacitors. Hybrid supercapacitors comprise the compensations of pseudocapacitors and EDLCs [19]. The limitations in both supercapacitor types are eliminated in the hybrid system and show better electrochemical characteristics.

Therefore, the present study is an attempt to give views to the researchers to widen their eyes to discover and develop flexible energy storage devices based on textile materials. The use of flexible supercapacitors undoubtedly brings new alternatives to the energy storage mechanisms. Efforts are underway to develop supercapacitors that are bendable, lightweight, and even wearable energy storage devices. Our review focused on pseudocapacitors as they do have higher energy densities and brought extraordinary capacitance behavior. The review paper only presents supercapacitors based on banana peel and three conductive polymers (PEDOT:PSS, PPy, and PANI) since they are highly explored and present high electrode performance based on our search. The selection was made on the performance and the applicability parameters which will be discussed in separate section

2. Supercapacitors Based on Carbonized Banana Peels

Banana peels are conventionally waste materials that habitually discarded after consuming the edible parts. This causes a temporary pollution to the surrounding area where high marketing areas are available. On the other hand, the depletion of fossil fuels along with the dynamic climatic alteration requires extra exploitation of spotless and justifiable energy alternative sources [8]. In addition, the recent surge of flexile and lightweight electronics with high durability pushes most industries to the implementation of the best alternatives to renewable energy sources. Among the flexible and lightweight energy storage devices, supercapacitors are widely researched. Supercapacitors are the most promising energy storage alternatives for short-term applications. Supercapacitors have high power density and fast charge-discharge rate beyond they do have long lasting life cycle against the surrounding environment [20].

Carbon-based porous structure materials have been proven to be promising as flexible, lightweight, and durable supercapacitors [20]. Porous supercapacitors based on carbonized banana peel is one of the promising research areas in recent years [9,21–23]. Banana peel can be prepared for a possible supercapacitor in different ways. Figure 2 illustrates one route of manufacturing supercapacitor from carbonized banana peel.

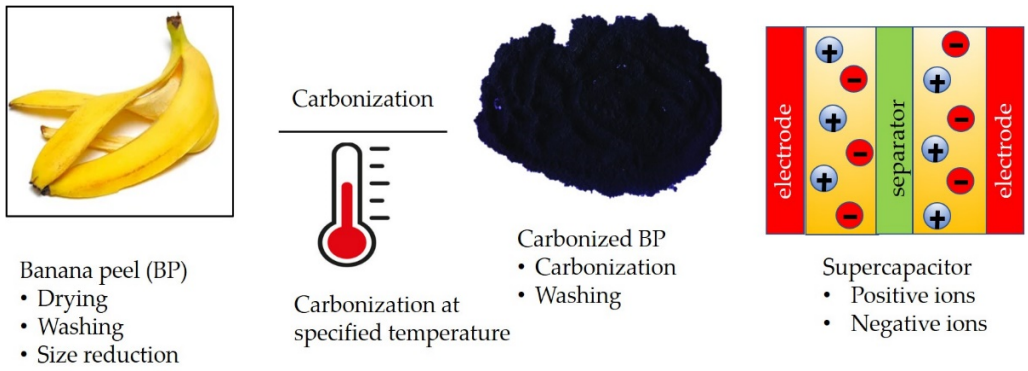

Figure 2. Illustrations showing process of manufacturing flexible supercapacitors starting from raw material preparation, carbonization and supercapacitor assembly.

Banana peel is an excellent potential for manufacturing supercapacitors. Various carbonization methods have been employed to improve the performance of supercapacitors with irrespective of its purposes and achieved high capacitance values. Table 1 summarizes the applied carbonization methods and its effect on the capacitance values.

Table 1. Summary of carbonized supercapacitor (SC) manufacturing based on banana peel.

Method of Carbonization	Purposes	Results Achieved	Ref.
One-step chemical activation	Improving electrochemical performance	Capacitance of 227 Fg^{-1} at 1 Ag^{-1}	[24]
Carbonization without activation	Green and cost-effective facile route	Notable specific capacitance (811 Fg^{-1})	[21]
one-step hydrothermal method	To get excellent electrochemical performance	Capacitance enduring 51 Fg^{-1} at 5.0 Ag^{-1}	[25]
Chemical activation	To improve conductivity and electrochemical performance	Specific capacitance of 90.23 Fg^{-1} at 10 mVs^{-1}	[26]
Heating banana peel soaked with KOH at high temperature	To check stability against multiple electronic cycling and bending	Devices displayed high areal capacitance of 88 mF/cm^2 at 10 mV/s scan rate	[27]
two-step hydrothermal process	To reach easily to the active site and to shorten the ion transport path	Large capacitance of 816 Fg^{-1} at the current density of 5 mA cm^{-2}	[28]
Green pyrolysis	To check energy storage ability and environmental remediation	Capacitance of 655 Fg^{-1} in 1 M at a current density of 0.35 Ag^{-1} & excellent cyclic stability of 79.3%	[29]
Sulfur-doped (chemical carbonization)	Sustainable supercapacitor production	High Brunauer-Emmett-Teller surface area of 2224.9 m^2/g, a large pore volume of 0.77 cm^3/g	[22]
Carbonization with chemical activation	To see relationship of surface area to cell capacitance	SC increased from 59–~265 Fg^{-1} at 0.1 Ag^{-1}	[30]
Biological activation	Optimization of precursors and synthesis methods	Get specific capacitance of 476 Fg^{-1} in 1 M H_2SO_4 electrolyte	[31]
Chemical co-precipitation method	To get high electrochemical property	specific capacitance of 465 Fg^{-1} at a scan rate of 10 mV s^{-1} by CV	[23]
Biological fermentation	Stabilize the structure of electrodes	High-capacity hold of 58.35% after 100 cycles	[32]
Hydrothermal method	To increase the electrochemical performance	Had a specific capacitance of 139.6 Fg^{-1} at 300 mA g^{-1}	[33]
straightforward carbonization	To improve electrochemical performance	Specific capacitance improved from 59 to 258–273 Fg^{-1} at 0.1 Ag^{-1}	[34]
KOH pellets at different carbonization temperatures	To improve electrochemical performance	Specific capacitance of 165 Fg^{-1} at energy density of 18.6 Wh kg^{-1} at 0.5 Ag^{-1}	[35]
Carbonization followed by activation	To see the relationship between surface area and electrochemical property	Surface area had significant effect on electrochemical property (specific capacitance of 68 Fg^{-1})	[36]
H_2SO_4 activation	Carbon nanofiber synthesis	Carbon nanofiber formed at 700 °C)	[37]
KOH	Absorption study	reflection loss peak of −44.59 dB at 10.84 GHz	[38]

As indicated in Table 1, carbonization of biomass resulted in carbon materials with porous structure and hence carbon materials have used for energy storage materials as supercapacitors [39]. Recent developments in the production of flexible supercapacitors indicated that banana peels represent a possible method to produce durable, flexible, and light weight energy storage devices. Supercapacitors based on banana peel achieved high capacitance value due to the ability of embodying high surface area which in turn can encompass highly porous structures. This most likely proves that carbon materials based on banana peels will continue to strive to be one of the protentional applications for

supercapacitor production as far as optimization in terms of activation type, high porosity, and flexibility achieved.

Banana peels (BP) have been used as the supercapacitor electrode after carbonization. They can be carbonized by applying various mechanisms such as chemical activation [40], hydrothermal methods [33], biological fermentation [32], and pyrolysis [29]. For the sake of obtaining higher capacitance value, various chemicals may be employed to chemically activate the banana peel carbon. The chemical activation may increase the surface area of the carbonized banana peel with affecting the structure of activated carbon. Activation helps to increases the surface area to volume ratio and to enhance absorption is enhanced. Moreover, the supercapacitor for chemically-activated banana peel achieved exceptional recurring steadiness with capacitance maintenance of ~97% for 5000 cycles [24]. Banana peel carbon provides the highest performance when activated using chemicals. Furthermore, the chemical precipitation has helped to enhance the supercapactive performance of carbonized supercapacitors, and hence chemical activation in suggested for the production of durable supercapacitors in the future.

3. Supercapacitors Based on PEDOT:PSS

3.1. Introduction

Poly(3,4-ethylenedioxythiophne)-polystyrene sulfonate (PEDOT:PSS) is the most recently explored, widely used, and successfully implemented intrinsically conductive polymers. The utmost plausible reason for this is due to excellent electromechanical performance [41], high conductivity and wash durability [42], and water processability and light transmissive behaviour [43,44]. In addition to this point, PEDOT:PSS is stable in environment and can be processed up to high temperature ranges. PEDOT:PSS alone is not good conductor as hydrophilic PSS hides the conductivity. In this case, some researchers have been using polyethylene glycol (PEG) [35], dimethyl sulfoxide (DMSO) [36], and ethylene glycol (EG) [37] to enhance the conductivity. When PEDOT:PSS is doped with these solutions, structural rearrangement has taken place (Figure 3c) [42].

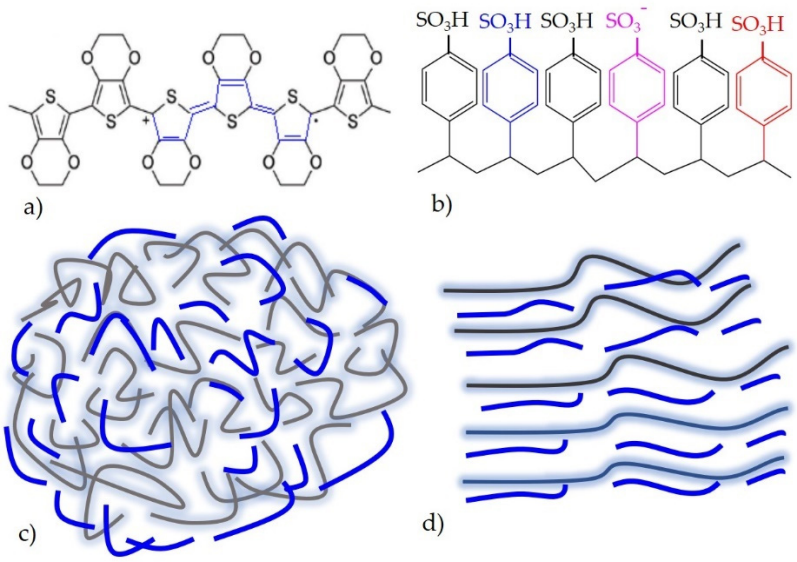

Figure 3. (**a**) Chemical structure of PEDOT; (**b**) chemical structure of PSS; (**c**) when PSS is added to PEDOT; a water dispersion solution is created; (**d**) when PEDOT:PSS is doped with conductive enhancers, a structural re-arrangement is formed, adapted from Ref. [42].

Perhaps the most well-known example of an organic intrinsic semiconductor polymer is the polymer poly(3,4-ethylenedioxythiophene) (PEDOT), predominantly when it intricates with poly(styrene sulfonate-PSS) (PEDOT:PSS). PSS surrounds and mostly found on the surface of PEDOT and which helps PEDOT to be dispersible in water. It is extremely conductive,

light-transmissive to a significant extent, water-processable, and highly flexible. Considerable recent research on this ubiquitous material has focused on enhancing its deformability beyond flexibility (a property that any suitably thin material possesses) to stretchability (a property that requires molecular or nanoscale structure engineering). The aforementioned properties of PEDOT:PSS are fundamental, which may help researchers to investigate the use of PEDOT:PSS as a durable energy storage materials such as supercapacitors.

Poly(3,4-ethylenedioxythiphene)-poly(styrene sulfonate) has been used in the production of high-performance supercapacitors. Table 2 summarizes the use of PEDOT:PSS to produce supercapacitor and its efficiency. Using PEDOT:PSS conductive polymer not only brought electrochemical performance but also possess excellent durability against washing [41].

Table 2. Summary of supercapacitors based on PEDOT:PSS with capacitance (C) main factor.

Precursor/Composite	Capacitance	Performance	Ref.
rGO	226.5 F cm^{-3}/279.3 mF cm^{-2} at 0.5 A cm^{-3}	74.7% C retention at 50 A cm^{-3}	[45]
WO_3	1139.6 mF cm^{-2} at 2 mA cm^{-2}	Working voltage of 1.6 V	[46]
Cellulose nanofibrils	854.4 mF cm^{-2} at 5 mV/s	Areal ED of 30.86 μWh cm^{-2}	[47]
PANI, PPy	156 mF cm^{-2} C at 1 mA cm^{-2} CD	41% capacity persisted at 20 mA cm^{-2}	[48]
$MgTf_2$	280 Fg^{-1} at 3 mV/s and 376.6 Fg^{-1} at 100 mA g^{-1}	PD ~100.08 Wkg^{-1}	[49]
Graphene	C of 2 mF cm^{-2} at a scan rate of 10^2 mV s^{-1}	>95% C retaining after 10^3 cycles	[50]
Polypyrrole	12.4–10.5 F cm^{-3} at a CD of 40–320 mA cm^{-3}	C retention rate of 88.1% for 10^3 charges/discharge cycles	[51]
Carbon nanofibers	C of 1321 Fg^{-1}, at a scanning speed of 1 mV/s	Retention of 80% of its performance after 2500 CV cycles	[52]
MnO_2 microspheres	Capacitance of 135.4 mF cm^{-2}	94% C maintenance after 3000 cycles	[53]
$rGO/CoFe_2O_4$	Capacitance of 229.6 mF cm^{-2}	ED and PD of 25.9 Wh kg^{-1} and 135.3 W kg^{-1}, respectively	[54]
Poly(acrylamide)	specific C of 327 Fg^{-1} at 3 mV/s	highest ionic conductivity of 13.7×10^{-3} S/cm at 22 ± 2 °C	[55]
CoCCHH-CoSe	C of 440.6 Fg^{-1} at 1 Ag^{-1}	ED of 137.7 Wh kg^{-1}	[56]
PANI Nanofiber	C of 301.71 mF cm^{-2} at CD of 1 mA cm^{-2}	ED of 0.023 mWh cm^{-2}, with PD of 0.279 mW cm^{-2} at a lower current density of 1 mA cm^{-2}	[57]
nanoflower MnOx	C of 580 mF·cm^{-2} at 0.5 mA	>90% for 40% stretch	[58]
—-	C of 3.92 mF/cm^2 at 1 mA/cm^2	C retention > 90% after 3×10^3 cycles	[59]
—-	Capacitance of 990 mF cm^{-2}	C retention of 74.7% after 14,000 cycles	[60]
Alginate/PPy	Capacitance of 246.4 mF cm^{-2}	97% of initial values after 180° bending	[61]
Ag-coated Tyvek	Mass C (138.7 Fg^{-1}) & volume C (544.2 F/cm^3) at the scan rate of 50 mV/s.	91.2% retention after 100 cycles	[62]
PVA/H_2SO_4	Areal C of 44.5 mF cm^{-2} at PD of 0.04 mW cm^{-2}	92% retention at 200% stretchability	[63]
MWCNT	specific capacitance of 235 Fg^{-1} at 5 mV s^{-1}	retention of about 92% in 1M H_2SO_4 electrolyte	[64]

GO, reduced graphene oxide; WO_3, tungsten trioxide; $MgTf_2$, magnesium trifluoromethanesulfonate; CD, current density; PD, power density; ED, energy density; CoCCHH-CoSe, heterogenous tube; MWCNT, multi-walled carbon nanotubes.

3.2. Conductivity Enhancement Principle to Increase Capacitance Value

It is well known that in the use of PEDOT:PSS in supercapacitor applications is due to its conductivity. Treating PEDOT:PSS with conductive enhancers has improved the supercapacitance efficiency [65,66]. Glycerol-dopped PEDOT:PSS [67] which has been coated on polyamide fabric has helped to fabricate flexible and wearable energy storage textiles. This method of fabrication of energy storage devices helps the user to produce user friendly garments which are light weight and can be worn. Human needs are demanding and dynamic in wearable garments. Being light weight, flexible, and washable are some of the requirements in wearable garments. Dong et al. [68] have tried to address these issues by developing portable, stretchable, and washable multifunctional wearable textiles based on PEDOT:PSS and ethylene glycol (dopant) solutions. The intended fabric has generated electric energy with a supreme immediate peak power density of ~85 $mW \cdot m^{-2}$ and light up at least 124 light-emitting diodes. This shows that it is conceivable to develop long stable textile materials with excellent capacitance value where the supercapacitance characteristics could be achieved well.

On the other hand, treating the substrate material with chemical solutions help to enhance the conductivity of the substrate and hence the supercapactive efficiency [69]. Zhang, Wei, et al. [70] claimed that treating textile materials with ethanol and doping of PEDOT:PSS with sodium dodecylbenzene sulfonate (SDBS) have brought an in increase in the conductivity of the materials so that supercapacitor performance improved. When PEDOT:PSS is treated with anionic surfactants, segment separation between PEDOT and PSS will occur, which means the hydrophilic PSS part will move towards the surface of PEDOT and the conductive PEDOT will diminish. Hence, the conductivity which was enhanced as a result of the performance of the material to be used as an electrode material will be increased. Furthermore, pretreatment of textile substate will allow easy diffusion of the conductive PEDOT so that high durability achieved. Pre-treating textile fabric with double barrier dielectric (DBD) atmospheric plasma in air has changed the property of the fabric completely [67] which further coated with PEDOT:PSS based solution to bring flexible heating element which can encapsulated into wearable garment.

Not only chemical solutions, but also nanoparticles based on V_2O_5 have increased the conductivity of PEDOT:PSS. Hence, the supercapacitance value has been doubled [71]. The addition of vanadium element brought further redox reactions and hence movement of electrons facilitated. Increasing conductivity by means of chemical solutions [72] will further enhance the supercapactive performance of PEDOT:PSS based supercapacitors. PEDOT:PSS based supercapacitor performances can be enhanced using composite materials based on carbon nanotubes [73,74], MoO_3 nanowire [75], titanium carbide [76], reduced graphene oxide [77], alginate hydrogels [78], tungsten oxide and gold nanoparticles [79], silver nanowires [80], polyvinyl butyral [81], molybdenum disulphide [82], cellulose nanofibrils [83], etc. The most reliable supercapacitors based PEDOT:PSS polymer can be obtained incorporating heterogenous materials with conductive polymers. Cost effective, easily processable, scalable, and high-performance supercapacitors can be produced ultimately.

In summary, rGO and nano-based materials are used commonly as a precursor material to further enhance the electrode performance of PEDOT:PSS based supercapacitors. In addition, additional conductive polymers can be combined with PEDOT:PSS to increase the specific capacitance of the materials. Excellent stretchability up to 200% has been achieved using sulphuric acid an electrolyte.

3.3. Textile Based Supercapacitors Using PEDOT:PSS

Flexible supercapacitors based on PEDOT:PSS using textile materials as a substrate base material are attractive for energy storage devices due to their low cost, flexibility, stretchability, lightweight characteristics, and excellent electrochemical properties [53,84,85]. Smart textiles are materials that can be worn on human body with a two-fold purpose; protection against the environment and additional functionality such as energy storage in smart textile applications [86]. Wearable supercapacitors are one of the self-powered smart

textile with sweat based supercapacitors with excellent washable behavior [10]. Figure 4 illustrates wearable electronics based on textile materials.

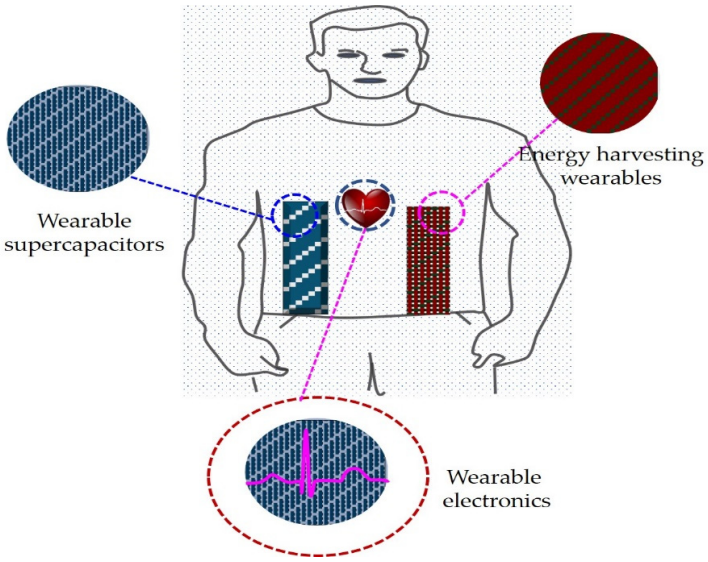

Figure 4. Personal and wearable electronics realized by introducing conductive polymers on wearable fabrics. The fabrication follows easy, simple and uses simple design.

The multifunctionality clothing are advanced toward an era when wearability and comfortability are the most important trends. Rapid growth of wearable textiles increases the demand of flexible power sources which have comparable comfort and stretchability during service. Cyclic stability and electrochemical performance are very important for fiber-based materials [87,88]. Textile-based supercapacitors are highly demanding due to ease to manufacture large-area electrodes [89] in addition to their flexibility and stretchability [89–93]. In textile-based supercapacitors, yarn twist has its own effect on the efficiency of the device [94] when durability for repeated bending is in action for the supercapacitor material. Adding composites such as polyaniline and manganese oxide on PEDOT:PSS conductive polymer on cotton textiles brought excellent capacity retention and can be used as disposable power sources for e-textile that can be worn in the human body [95]. PVA (polyvinyl alcohol)–PEDOT:PSS hybrid deposited on carbon nanotube (CNT) yarn [93] to create flexible, lightweight, and wearable high performance supercapacitors. In general, textile-based supercapacitors are easy for fabrication, low-cost, sustainable, disposable, and easily manageable for wearable electronics applications.

In summary supercapacitors based on PEDOT:PSS showed superior performance in addition to its durability and compatibility. Furthermore, doping PEDOT:PSS with ionic solution will further enhance the conductivity of the PEDOT:PSS and consequently, the performance of the supercapacitor is improved. Therefore, it could be recommended that flexible textile-based supercapacitors can be developed effectively using PEDOT:PSS as an electrode material.

4. Supercapacitors Based on Polyaniline

Polyaniline(PANI) is considered as one of most promising and versatile conducting polymers, and has been studied extensively as an electrode or super capacitor device due to its high capacitance, synthesized easily, inexpensive, high electro activity, high doping level, excellent stability, high specific capacitance (400–500 Fg^{-1} in an acidic medium), good environmental stability, controllable electrical conductivity (around 0.1 S cm^{-1} in the doped state with a Li dopant, but can range from around 0.1 to 5 S cm^{-1}, easily processed, wide capacity range from 44 to 270 mAh g^{-1} [96–98]. However, PANI also has its drawbacks such as volumetric swelling and shrinking at the time of charge and discharge as a result of ion

doping and de-doping, this volumetric variation destroys the backbone of PANI resulting lifecycle weakening. This drawback can be solved by making PANI based composites of PANI with carbon based materials [97].

Titanium nitride nano wire array surface was coated by carbon and PANI sequentially to produce shell/shell/core polyaniline/carbon/titanium nitride nanowire composite, carbon and PANI as a shell and titanium nitride as core, to be used as electro active electrode material for super capacitor application by researcher Xie, Y. et al. [99]. The study reveals that the composite exhibited 1093 Fg^{-1} specific capacitance at 1.0 Ag^{-1}, good cycling stability with 98% capacity retention after 2000 cycles. Gupta, V. and Miura, N. [100] have deposited PANI onto single-wale carbon nanotube by electro chemical polymerization to produce PANI/single-wale carbon nanotube composites used as super capacitor having specific capacitance of 485 F/g.

Rajkumar, S. et al. [101] have made the synthesis of porous nano rod-like structure $FeCo_2O_4$/PANI composite electrode by in-situ polymerization method applicable as super capacitors. Also a highly ordered 3D α-Fe_2O_3@PANI core-shell nanowire arrays was fabricated by Lu, X.-F. et al. [102]. The produced core-shell composite has higher reaction surface area, fast ion and electron transfer and good structure stability. PANI was deposited on crystalline α-MoO_3 surface via in-situ polymerization at room temperature that gives a high performance MoO_3/PANI supper capacitor with a specific capacitance of 714 Fg^{-1} at 1 mV s^{-1}.

Srinivasan, R. et al. [103] synthesized a potential electrode for Co-MOF/PANI composite was from cobalt nitrate, 1,3,5-benzene tricarboxylic acid and aniline via in-situ chemical oxidative polymerization technique with specific capacitance of 504 Fg^{-1} at of 1 Ag^{-1} current density which can sustain 90% of its initial capacitance up to 5000 GCD cycles at current density of 2 Ag^{-1}. PANI and manganese dioxide (MnO_2) synthesized on carbon cloth (CC) by electrochemical polymerization with the addition of $LiClO_4$ to increase the PANI/MnO_2 electrode.

The energy used by supper capacitors cannot be recognized until the energy of super capacitors refuse. To solve this problem, a promising smart super capacitor such as super capacitors based on PANI that can change color at different potential (0–1 V) s. As the device shows different colors at different potential with visualized energy levels, we can know the energy consumed out before the device stops. Inamdar, A.I. et al. [104], also fabricated smart super capacitor nickel oxide-PANI electrodes by pulse potential electrodeposition method. The electrode exhibited specific capacity of 539 Fg^{-1}, change color at different stored energy level (dark blue when the charge is fully charged, transparent when fully discharged, and intermediate colors). Table 3 summarizes supercapacitors-based o PANI.

Table 3. Supper capacitors devices or electrodes based on PANI.

No.	Super Capacitor Electrode or Devices Based on PANI	Method of Manufacturing	Capacitance Value	Ref.
1	PANI	chemical oxidative polymerization	267 Fg^{-1}	[104]
2	H_2 bonded graphene/PANI	chemical oxidation	598 Fg^{-1} at 1.0 Ag^{-1}	[105]
3	PANI/carbon/titanium nitride nanowire composite	sequentially coating carbon and PANI on the surface of TiN Nano wire	1093 Fg^{-1} at 1.0 Ag^{-1}	[99]
4	PANI/graphene	Chemical vapor deposition	789.9 Fg^{-1} at 10 mVS^{-1}	[106]
5	PANI/nanowire	Spin-coated	Areal capacitance of 0.017 Fcm^{-2} at 5 mV S^{-1}	[107]
6	PANI/SWCNT composites	electro chemical polymerization of PANI onto SWCNTs	485 Fg^{-1}	[100]

Table 3. *Cont.*

No.	Super Capacitor Electrode or Devices Based on PANI	Method of Manufacturing	Capacitance Value	Ref.
8	PANI/C-TiO_2 NTAs	Ar atmosphere	104.3 mF cm^{-2}	[98]
9	PANI)/carbon aerogel	chemical oxidation polymerization	710.7 Fg^{-1}	[108]
10	$Ni_3V_2O_8$@PANI composite	in situ chemical bath	2565.7 Fg^{-1} at 5 mV/s	[109]
11	PANI/Co-Porphyrins composite	——	823 Fg^{-1} at 0.5 Ag^{-1}	[110]
12	hollow Co_3O_4/PANI Nano cages	in situ surface polymerization	1301 Fg^{-1} at CD of 1 Ag^{-1}	[111]
13	MoO_3/PANI	in situ polymerization	632 Fg^{-1} at a CD of 1 Ag^{-1}	[112]
14	Ni-PANI film electrode	Multi-step electrode position	543 at 1 Ag^{-1}	[107]
15	PANI/graphene oxide composite	in situ polymerization of aniline monomers in the presence of GO	206 Fg^{-1} at 1 Ag^{-1}	[113]
16	graphene oxide–polyaniline	in situ polymerization	525 Fg^{-1} at 0.3 Ag^{-1}	[114]
17	Co-MOF/PANI composite	coupling	162.5 Cg^{-1} at 0.4 Ag^{-1}	[115]
18	carbon cloth/PANI-MnO_2	electrochemical polymerization	634.0 Fg^{-1} at 1 Ag^{-1}	[116]
19	PVA/carbon nanotubes/PANI film	in situ polymerization of PANI on the surface of PVA/CNT films	196.5 mF cm^{-2}	[117]
20	reduced graphene oxide/Zn-Metal-organic frameworks@PANI	in situ polymerization	372 Fg^{-1} at 0.1 A g^{-1}	[118]
21	Honeycomb-like nitrogen-doped carbon/PANI composite	in situ polymerization	686 Fg^{-1} at 1 Ag^{-1}	[119]
22	brannerite type copper vanadate/PANI	in situ polymerization	375 Fg^{-1} at 4 Ag^{-1}	[120]
23	CNF/thionickel ferrite/PANI ternary nanocomposite	in situ polymerization	645 Fg^{-1} at CD of 1 Ag^{-1}	[121]
24	manganese sulfide/graphene oxide/PANI nanocomposite	in situ polymerization	822 Fg^{-1} at 10 mV/s	[122]
25	PANI/Boron carbo nitride nanocomposite	in situ polymerization	67.1 Fg^{-1} at a scan rate 5 mV S^{-1}	[123]
26	PANI/MIL-101	as-synthesized	1197 Fg^{-1} at 1 Ag^{-1}	[124]
27	PANI/perlite-barium ferrite nanoparticles composite	hydrothermal	330 Fg^{-1}	[125]
28	CuCe-bimetal organic frameworks@PANI-1	hydrothermal	724.4 Fg^{-1} at 1 Ag^{-1}	[126]
29	PANI-graphene/PVA/PANI-graphene	chemical activation	1412 Fg^{-1}	[127]
30	PANI/Ag@MnO_2	deposition	1028.66 Fg^{-1} at 1 Ag^{-1}	[128]
31	PANI/*p*-phenylenediamine—GO composites	in situ polymerization	635.2 Fg^{-1} at 1 Ag^{-1}	[129]
32	GO/MnO_2/PANI nanocomposites	polymerization	150 Fg^{-1}	[130]
33	3D graphene oxide/PANI-carbon fiber paper	template method	1013 Fg^{-1} at 1 Ag^{-1}	[131]
34	rGO/unzipped CNT/PANI	in situ polymerization	359.3 Fg^{-1} at 1 Ag^{-1}	[132]
35	Multi-growth site graphene/PANI composites	oxidation	912 Fg^{-1} at 1 Ag^{-1}	[133]

SW/CNT, single walled/carbon nanotube; r/GO, reduced/graphene oxide; CD, current density; CNF, carbon nanofibers; MOF, metal-organic frameworks; NT, nanotube.

As indicated in Table 3 the supercapacitor devices or electrodes produced based on high conducting PANI polymer either by electro polymerization technique, coating or other methods. In summary, conductive polymers, in this case PANI, are the basis for supercapacitance production.

In conclusion, PANI based supercapacitors as an electrode is highly recommendable to produce flexible and rollable energy storage devices. In addition, using various composites is uphold to further enhance the super capacity performance.

5. Supercapacitors Based on PPy

In the last few years, portable, light weight, flexible (stretchable), and wearable supercapacitors are more researched due to the increase in the demand of the human being. Supercapacitors also called electrochemical capacitors, can store electrical energy by means of either ion adsorption (electrochemical double layer capacitor) or fast surface redox reactions (pseudo capacitors). A supercapacitor device contains four elements: electrode, electrolyte, current collectors, and membrane. The electrode materials are the key element in supercapacitors with large surface area and conductivity properties [134].

Among the conductive polymers that can be used as an electrode supercapacitors to store chemical energy via redox reaction are polypyrrole (PPy), polyaniline (PANI), and poly(3,4-ehtylenedioxythiophene)-(PEDOT), used as stretchable supercapacitor electrode materials with the required performances which have great importance in wearable electronic systems [135]. PPy is a pseudo capacitive material having good conductivity, high specific capacitance [135–138], good biocompatibility, environmental friendly [139], electroactive, light weight, highly flexible, low cost [135,140], and can be used as a potential flexible electrodes candidates for wearable flexible super capacitors [141,142].

On the other hand, the electrode based on pure PPy structure has its own drawbacks such as poor stability issues during charge/discharge cycles due to the repeated doping and doping/de-doping that results in the PPy molecular skeleton to be repeatedly extend and shrinkage leading the material to crack, and cyclic stability deterioration [135]. Synthesis of composites PPy with other polymers including nanofibers, graphene, CNT, carbon, etc. [143–151] can enhance the performance of supercapacitors. The other solution to solve these drawbacks is to mix PPy with pseudo capacitive metal oxides of RuO_2, MnO_2, two-dimensional materials of MoS_2, carbonitrides [152], electric double-layer capacitive type nanostructured carbon materials [153,154], carbon nanotubes, and biomass-derived carbon [155]. This brought an excellent cyclic stability and withstand the crack caused.

Mixing of PPy polymers with other pseudo materials has also its own limitations such as process complexity, high cost and insignificant cyclic performance improvements. As a result, researchers have tried to solve the super capacitive performance limitations by combining PPy with nanostructured materials with a unique high-surface-area by electro polymerization technique such as PET/reduced graphene oxide/PPy composite electrode [139], PPy-multi-walled-carbon-nanotube-silk [140], carbon-nanotube/PPy composite [131], and successful construction of sea urchin spines-like PPy arrays on cotton-based fabric electrode via a facile electropolymerization and employ it in enhancing the cyclic stability of the supercapacitors [143].

Conductive polymer-based on fabric electrodes are used as a flexible electrode substrate which are used for the production of supercapacitors due to their positive characteristics such as comfort, light weight, low-cost, water absorption, and excellent mechanical strength. Such flexible electrode materials include PPy/reduced graphene oxide nano composite cotton fabric [134], EPPy-PPy/Poly-(vinyl alcohol-co-ethylene) nanofibers/cotton fabric [137], polyethylene terephthalate/reduced graphene oxide/PPy [139], PPy/carbon cloth [142], and PPy-carbonitrides coated textile [132].

The summarized research works that are based on PPy and its composite electrodes for supercapacitors applications, and their investigations are presented in detail (Table 4).

Table 4. Summary of supercapacitors based on PPy in terms of capacitance retention (CR), specific capacity (SC), energy density (ED), areal capacity (AC), power density (PD), and capacitance (C).

No.	PPy Based Electrode/ Super Capacitor	Method of Manufacturing	Investigated Properties	Values	Ref.
1	EPPy-PPy/CF	electropolymerization	SC	617.5 mF cm^{-2} at 0.4 mA cm^{-2} CD	[143]
2	PPy/CNOs	template-degrading	SC	64 Fg^{-1}	[144]
3	PET/Reduced graphene oxide/PPy composite electrode	oxidation polymerization	AC; VC; ED; PD and RC after 6000 cycles	0.23 cm^{-2} at a scanning rate of 1 mV s^{-1}; 5.5 F cm^{-3} at a discharge CD of 1.6 mA cm^{-3}; 11 mWh cm^{-2}; 0.03 mW cm^{-2} at 6.86 mg c^2; 76%	[145]
4	PPy-Multi-Walled Carbon Nanotube-silk electrode	polymerization	SC; CR, after 3000 cycles	676.9 mF cm^{-2} or 376.3 F cm^{-3}; 81%	[146]
5	PPy/reduced graphene oxide Nano composite cotton fabric	chemical polymerization	SC; CR after 10^4 cycles	9300 m^{-2} at 1 mA cm^{-2}; 94.47%	[140]
6	Fabric based polyethylene terephthalate/reduced graphene oxide/PPy	dipping and drying	SC; CR after 6000 cycles; ED; PD	230 mF cm^{-2} at 1 mV s^{-1}; 76%; 11 µWh cm^{-2}; 0.03 mW cm^{-2}	[145]
7	PPy-Cotton electrode	In situ polymerization	Specific capacitance	268 Fg^{-1} at a scan rate of 5 mV s^{-1}	[134]
8	PPy-Viscose rayon electrode	In situ polymerization	Specific capacitance	244 Fg^{-1} at a scan rate of 5 mV s^{-1}	[134]
9	Parallel CNT/PPy composite	electro chemical deposition	Specific capacitance	139.2 Fg^{-1} (27.8 mF cm^{-2}, 10 mV s^{-1})	[137]
10	Twisted carbon nanotube/PPy composite	electro chemical deposition	Specific capacitance	331.4 Fg^{-1} at 5 mV s^{-1}	[137]
11	PPy@ acid-pre-treated stainless steel yarn electrode	electro chemical deposition	VC; ED; CR at 6000 cycles	14.69 F cm^{-3} at CD of 25 mA cm^{-3}; 3.83 mWh·cm^{-3} at a PD of 18.75 mW cm^{-3}; 90%	[147]
12	PPy-carbonitrides coated textile electrode	dipping and drying	SC; ED; PD	343.20 Fg^{-1}; 1.30 mWh g^{-1}; 41.1 mW g^{-1}	[138]
13	PPy/carbon cloth electrode	electro chemical	Areal specific capacitance	174.5 mF cm^{-2} at scan rate of 5 mV s^{-1}	[148]
14	vanadium pentoxide/functionalized CNT/PPy composite electrode	VP polymerization	AC; CR after 10^3 charge-discharge cycles	1266 cm^{-2} at a CD of 1 mA cm^{-2}; 83%	[149]
15	PPy nanotubes/carbon cloth coated electrodes	interfacial polymerization	AC; CR after 500 cycles	0.74 F cm^{-2} at constant discharge & CD of 10 mA cm^{-2}; 79.5%	[150]
16	PPy/graphene nanoplatelets electrode	interfacial polymerization	AC	250 mF cm^{-2}	[151]
17	pristine polypyrrole membrane electrode	MO-assisted polymerization	CR after 1000 cycles; SC	88.9% cyclic stability; 509.8 Fg^{-1} at 0.5 Ag^{-1}	[152]
18	Paper derived activated carbon and bare/NF@PPy	hydrothermal & chemical polymerization	SC; ED	658 Fg^{-1} at a CD of 1 Ag^{-1}; 27.4 Wh kg^{-1}	[153]

Table 4. *Cont.*

No.	PPy Based Electrode/ Super Capacitor	Method of Manufacturing	Investigated Properties	Values	Ref.
19	porous PPy scaffold/conductive Cu_3 (2,3,6,7,10,11-hexa hydroxyl triphenylene)$_2$ catecholate electrode	polymerization	PD; ED; CR after 5000 cycles	233 mF cm^{-2}; 1.5 mW cm^{-2}; 12 µWh cm^{-2}; 85%	[154]
20	Cerium vanadate/PPy electrode	hydrothermal	SC; CR after 10^4 cycles	1236 Fg^{-1} at CD of 0.75 Ag^{-1}; 92.6%	[155]
21	PPy/sulfonated poly(ether ketone)/MWCNT electrode	In situ chemical oxidation	SC	593 Fg^{-1} at scan rate of 2 mV/s	[156]

MW/CNT: multiwalled/carbon nanotube; CNOs: carbon nanotube onions; VP: vapor phase; MO, methyl orange; CD, current density.

In general, PPy-based supercapacitors are promising candidates for the production cost-effective, simple, flexible, lightweight, wearable energy storage devices. When PPy is mixed with other materials such as graphene and nanomaterials, the specific capacitance value is enhanced. Furthermore, the cyclic stability and environmental durability also improved when PPy is mixed with other compounds such as cerium vanadate.

6. Conclusions and Future Perspectives

In conclusion, flexible, portable, lightweight, and wearable textile-based supercapacitors are highly demanding even though there still unsolved problems in many aspects. Different modalities exist depending on the characteristics of the base (supercapacitor) materials and the targeted energy storage materials. In all aspects, the performance of the supercapacitor, durability, and final application power rangers need to be fulfilled. The most commonly used materials for flexible supercapacitors are intrinsically conductive polymers such as PEDOT:PSS, PANI, and PPy. Furthermore, carbonized banana peels and their composites have been used for the production of high-performance supercapacitors. However, washability, durability against the environment, and electromechanical performance are the basic challenges of these types of supercapacitors. Therefore, a systematic approach of doping polymers, carbonization types, and pre-treatments are seen to offer opportunities for enhancing durability, efficiency, flexibility, and supercapacitor performance. However, most research conducted on textile-based flexible supercapacitors thus far has focused on their application as energy storage devices. Therefore, this review aimed to summarize the current state of research on textile-based flexible supercapacitors specifically developed for energy storage and harvesting and highlight research gaps to be addressed in future work.

It was found that flexible and textile-based electrodes have been produced with different material compositions and techniques. Even though no leading materials have been found the literatures, intrinsically conductive polymers such as PEDOT:PSS, PANI, and PPy appears to be the prominent solutions for the production of rollable and flexible supercapacitors. Furthermore, carbonized banana peels were the most researched material for a green and sustainable method for producing bendable and lightweight supercapacitors.

Sustainability is another issue that has rarely been addressed in textile electrode research. Sustainability issues are predominantly appropriate in the wearable textile segment, which involves the incorporation of numerous types of materials into a single system, thereby complicating durability, washability and resistance against the environment. Among others, significant features that should be considered include material choice, method of doping, pre-treatment selection in relation performance, durability, and en-

vironmental impacts. Therefore, future research should consider different as aspects of producing flexible, stretchable, long standing, and light weight supercapacitors.

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