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Paper

Flexible Energy-storage Devices: Maneuvers and Intermediate Towards Multi-functional Composites

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ABSTRACT: Flexible energy-storage devices (FESDs) have been studied and developed extensively over the last few years because of demands in various fields. Since electrochemical performance and mechanical flexibility must be taken into account together, different framework from composition of conventional energy-storage devices (ESDs) is required. Numerous types of electrodes have been proposed to implement the FESDs. Herein, we review the works related to the FESDs so far and focus on free-standing electrodes and, especially substrate-based ones. The way to utilize carbon woven fabric (CF) or carbon cloth (CC) as flexible substrates is quite simple and intuitive. However, it is meaningful in the point of that the framework exploiting CF or CC can be extended to other applications resulting in multifunctional composites. Therefore, summary, which is on utilization of carbon-based material and conductive substrate containing CF and CC for ESDs, turns out to be helpful for other researchers to have crude concepts to get into energy-storage multi-functional composite. Moreover, polymer electrolytes are briefly explored as well because safety is one of the most important issues in FESDs and the electrolyte part mainly includes difficult obstacles to overcome. Lastly, we suggest some points that need to be further improved and studied for FESDs.

Key Words: Flexible energy-storage, Flexible electrodes, Carbon fabric, Solid-state polymer electrolytes, Multi-functional composite

1. INTRODUCTION

Flexible energy-storage devices (FESDs) have been attracted intensive attention due to their potential applications such as wearable and portable electronics, sensing systems and devices for health care [1-3]. Researchers have generally implemented flexibility of FESDs by using electrodes where active materials are on the flexible current collectors (FCCs) such as carbon woven fabric, carbon or metal-coated plastic fibers, metal foils, conductive paper [1-4]; there also have been attempts to implant electrochemical functions into the FCCs (free-standing) [4,5]. In addition to these electrodes, polymer electrolytes are essential to construct FESD. For the lithium ion batteries (LIBs), liquid electrolytes have inherent instability induced by leakage and flammable solvents [6]. On the other hand, some researchers have made a shape conversion of the whole system while maintaining components of conventional LIB's electrode that consists of pulverized active materials with binder on the metal foil [7,8]. This approach has FESDs be easily employed practice applications. In the other, some have been realized FESDs from the yarn level as well as cable types [9-11]. In this, we focus on free-standing electrodes and, especially the ones on textile substrates that have flexibility and electrical conductivity concurrently as current collectors depending on what material they are made of. And, polymer electrolytes, which have risen as alternatives to liquid ones, were also briefly investigated.

When we figure out energy-storage devices (ESDs), it means something that transforms electrical energy to chemical one through a difference of electrochemical potential between two electrodes and vice versa. ESDs have several storing mechanisms such as intercalation/de-intercalation, electrical double layers capacitance (EDLC), and pseudo-capacitance; batteries generally carry not only the intercalation/de-intercalation but

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also conversion and alloy reactions [12-18], and supercapacitors operate with the EDLC and the pseudo-capacitance [19-21]. The larger power densities are required in industry, the more spotlights move to the supercapacitors that has moderate values in both energy and power densities. In spite of this circumstance, particularly lithium-based battery, which carries high energy density compared with other ESDs, still play important roles in FESDs [22-24]. It is difficult to tell which one is more momentous than the other is, especially in the FESDs. Therefore, regardless of the mechanisms, we went through free-standing, and substrate-based flexible electrodes (FEs) and full-cell composites in a viewpoint of energy-storage. The diverse carbon-based materials, such as carbon nanotube and graphene have been applied to the free-standing electrodes. Since these materials usually have excellent conductivity, they can be utilized as not only active materials but also substrate themselves. Furthermore, the mixture of the carbon-based and other active materials have been considered as promising candidates for FEs. The most intuitive way to manufacture FESD would be to use FCC. There have been endeavor exploiting conductive and non-conductive substrates. In addition to this, we suggested some factors that need to be further studied and improved for implementing commercially realizable FESDs.

2. FLEXIBLE ELECTRODES

2.1 Free-standing flexible electrodes

There are four main components in the ESD: electrode active materials, an electrolyte, a separator, and current collectors. Conventionally, copper and aluminum foils are exploited for current collectors of anode and cathode part respectively. However, the active material and the current collector can be combined into a free-standing FE overspreading conductive-layer on it if enhancement in conductivity is needed. And, carbon materials, such as carbon nanotube (CNT) and graphene, are commonly used to make the electrode due to its excellent conductivity, facile processes to give itself porous, chemical stability, and mechanical strength.

Zheng *et al.* [25] reported that the interconnected composite film, which is made of PEDOT:PSS, active nanoparticles, and CNT as shown in Fig. 1a, is one of the strategies to achieve FEs, especially superior in areal energy density. The composite was stable after 500th bending cycles; uniformly dispersed and interconnected CNT give the electrode flexibility to endure normal bending situations. Furthermore, since any active materials, which may be in the form of nanoparticles, can replace the material they used, we could easily modify the electrode in this framework. The CNT is likely to agglomerate one another to form film-type FE. For the LIBs, active materials, which possess high theoretical energy density like V_2O_5 [26-28], construct composite with CNT resulting in outstanding energy density with rate capability [29,30]. Wang *et al.* [31] made graphene paper coated with polyaniline (PANi) that is conducting polymer (Fig. 1b). After they manufactured the graphene film via filtration, they conducted electro-polymerization with the solution where the PANi monomer was dissolved in acidic environment of H_2SO_4. The synthesized SC had specific capacitance of 233 F g^{-1} and mechanical tensile strength of 12.6 MPa. As they mentioned, there is a trade-off between mechanical strength and electrochemical capacitance. The porosity is one of the key points in both properties; the more porous thing is better with respect to capacitance, whereas it can alleviate mechanical strength. According to applications, the two properties should be balanced appropriately, especially for this kind of free-standing electrode whose substrates are substaintially activated. Shiyong *et al.* [32] conducted carbonization to transform a melamine foam to a nitrogen-doped carbon foam that is conductive and low-cost. After that, the sample was hydrolyzed in boiling water (Fig. 1e). When all the procedures finished, it was checked for morphology of it to maintain its initial shape of foam as shown in Fig. 1c. In addition to this, it has porous structure as well (Fig. 1d). The synthesized foam was resilient enough to be bent

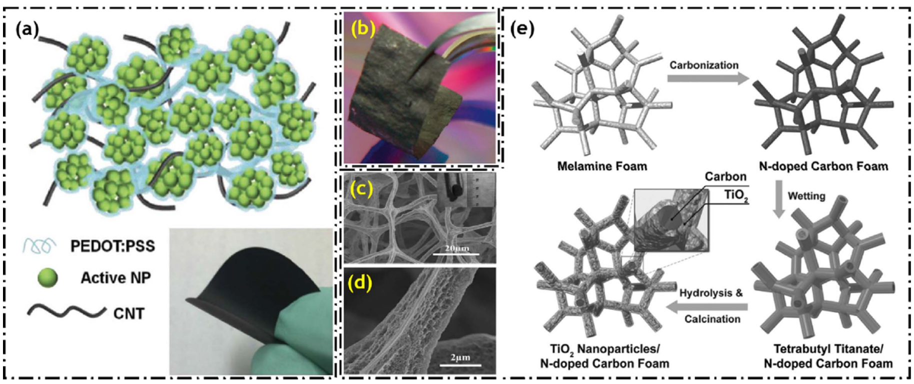

Fig. 1. (a) Schematic of active material, CNT and PEDOT:PSS composite from ref [25], (b) free-standing graphene paper from ref [31], (c) SEM image of N-doped carbon foam with TiO_2, (d) the magnified image, (e) Schematic of synthesis of the N-doped carbon foam with TiO_2 from ref [32]

reversibly with capacity of 188 mAh g^{-1} at 200 mA g^{-1}. The combination of TiO_2 and carbonized conductive frame facilitated outstanding capacity. However, although free-standing FEs have advantages in energy density by making whole system lighter, the weakened mechanical properties of the electrodes are inevitable because of the trade-off as mentioned before.

2.2 Flexible electrodes on substrates

The mechanical strength itself does not need to be taken into accounts in flexibility. However, in the perspective of bendable cyclic stability, using appropriate substrates could prevent performance degradation from excessive strain of the substrate. Additionally, since functions of active material and substrate are separated, we do not necessarily increase electrochemical performance at the cost of the properties of substrate. Furthermore, with the same electrodes, other types of multi-functional composites can be realized utilizing substrate's own strength [33-35]. The way to figure out FESDs would be simply using flexible conductive textile as current collector with appropriate active materials. Thereby, the FEs based on various textiles have been widely studied. Because the conductivity is necessary for substrate, researchers have used non-conductive textile with some conductive coatings on that and conductive textile like carbon cloth and carbon woven fabric to satisfy both flexibility and conductivity [36]. Some carried out conducting coating on plastic textile with metals or conducting polymer like PPy and PANi. And, CC and CF primarily gathered enormous attention because of their moderate conductivity and good mechanical strength with respect to weight [37]. Moreover, CC and CF have been steadily considered as promising candidates for applications as ESDs since the stacked-up shape of general carbon fiber reinforced polymer (CFRP) composites is similar to the sandwich-like structure of ESDs. In this structure, in which CC and CF were given as substrates and current collectors, mechanical properties like flexibility and load-bearing are almost solely contributed to the substrates while active materials on the conductive substrates play a role in electrochemical performance. It has especially a benefit in specific capacity because no more conductive additives are required unlike conducting coating of non-conductive substrates. However, the materials, which can be used in conductive substrates, are a few such as CC, CF, and metal meshes that could be not favorable to flexibility. On the other hand, the routes to exploit non-conductive textile as basic substrates can be easily employed to many kinds of textile like clothes even at the cost of a little loss in capacity with respect to weight.

2.2.1 Non-conductive substrates

Hu *et al.* [38] made cotton fabric conductive by a facile dipping method. They prepared ink solution where single-walled carbon nanotubes (SWNTs) are dispersed with the aid of surfactant (Fig. 2a). Then, they dip the textile into the solution and dried it several times resulting in conductive textile. Then, they deposited MnO_2 for further capacitance and mass loading in a given area (Fig. 2b, c). They said that functional groups on the textile and edge of SWNTs manifest strong van der Waals forces and hydrogen bonding between them ensuring strong attachment. According to this statement, this method is available for the textiles that have functional groups on them appropriately. We could also guess that further surface treatments, like O_2-low temperature plasma [39], increase a sort of physical and chemical bonds between SWNTs and given textiles with opening latent applications of various textiles.

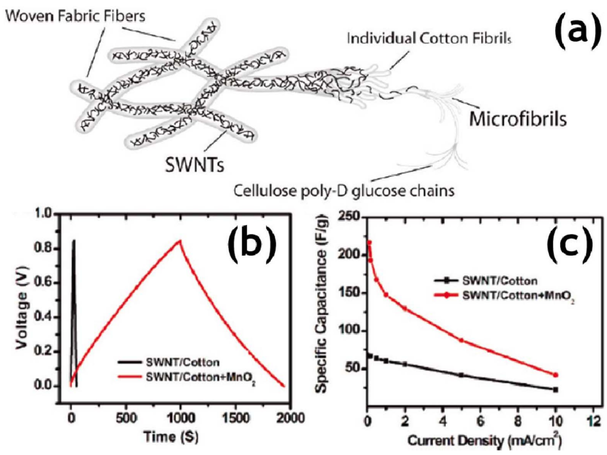

Fig. 2. (a) Schematic of cellulose fibers wrapped by SWNTs, (b) charge-discharge of the SWNT/cotton and the one with MnO_2, (c) Specific capacitance with respect to different current density from ref [38]

Lihong *et al.* [40] also used cotton fabric for a flexible SC. They immerse the cotton fabric, which is cut off from a conventional T-shirt as shown in Fig. 3a, in 1 M NaF solution followed by drying at 120°C for 3 h. Then, the textile was thermally cured in the tube furnace at around 1000°C. Through this manner, they transformed normal cotton fabrics to the activated carbon textiles (Fig. 3b, c). Moreover, MnO_2 was electrochemically deposited onto the textiles (Fig. 3d, e). They achieved approximately capacitance of 153 F g^{-1} at scan rate of 10 mV s^{-1} in the cyclic voltammetry (CV) test. In the point that they did not use any additional carbon material like CNT, graphene, and fullerene, the synthesized electrode is cost-effective with usage of MnO_2 that is relatively inexpensive than other active materials having pseudocapacitance. And, it is flexible like membrane that cannot support moment.

Yuxi *et al.* [41] functionalized graphene oxide (FGO) with hydroquinone by simply mixing and heating. They demonstrated their FGO gel electrode has remarkable capacitance through pseudocapacitance, and the flexible cell on polyimide substrates that were gold-coated for conductivity (Fig. 4a, c). The hydrogel electrode was pressed to make it flat, never-

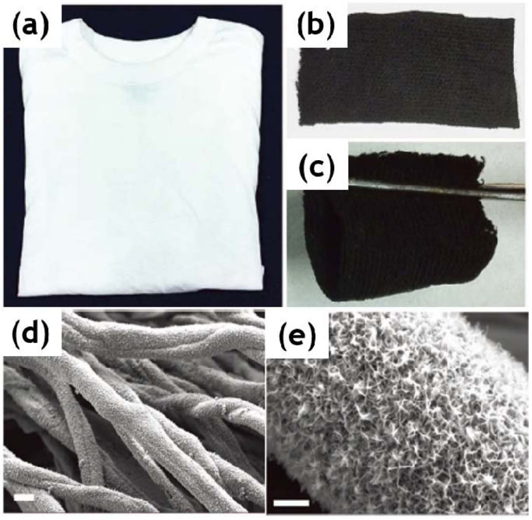

Fig. 3. (a) A commercial cotton T-shirt, (b, c) activated carbon textiles (ACT), (d, e) SEM images of the ACT coated with MnO_2 from ref [40]

theless, inner porous structure does not collapse (Fig. 4b). The H_2SO_4 and polyvinyl alcohol (PVA) gel was used as an electrolyte and a separator concurrently. They proposed that large surface area of graphene, beneficial porous graphene framework for ionic and electron transport, and superior pseudocapacitance by hydroquinone, which is attached onto graphene strongly via π-π interaction, contributes to outstanding electrochemical performance of the FGO. They demonstrated that proper combination between active materials and appropriate substrates facilitates FESDs.

Nanostructure manifest some merits in electrochemical reactions. First, high surface area makes reactions occur more frequently in many locations. Second, ionic diffusion path is shortened due to its nanoscale-length structure facilitating enhanced rate-capability. For these reasons, there have been some attempts to grow active materials on substrates directly. Nagaraju *et al.* [42] grew Ni-Co layered nanostructures on a textile substrate, which is originally copper-doped PET fibers as shown in Fig. 4d-h, by using electrochemical deposition. According to applied voltage, it showed different tendency in the formation of hydroxyl ions adjacent to the surface of the textile. For the specific voltage induced uniformly covered Ni-Co nanosheets on the conductive textile. The synthesized FEs have remarkable capacitance of 2105 F g^{-1} at current density of 2 A g^{-1}. The conductivity of the electrode itself is significant factor to determine the electrochemical performance. And, most of transition metal oxides have poor conductivity. Thereby, instead of transition metal oxides, use of metal sulfides is more favorable in the conductivity because of a smaller band gap. Jian *et al.* [43] proposed making textiles silver-sputtered to be employed as flexible conducting substrates. In addition to this process, they synthesized two kinds of ternary metal sulfides ($FeCo_2S_4$ and $NiCo_2S_4$) on the textiles. These metal sulfides conduct faradaic redox reactions interchanging sulfides to either hydroxides or oxides during the charging and discharging. The asymmetric SC (ASC) manifested good electrochemical performance of 1519 F g^{-1} at 5 mA cm^{-2}. Furthermore, it is noteworthy that almost all textiles can transform into conductive substrates by sputtering even though it can cause some problems in the ion batteries depending on operating voltage and metal that is sputtered. Sponges are also used as substrates due to its innate large surface area [44,45]. Chen *et al.* [45] used a sponge for a flexible sponge hybrid electrode. The synthesis route is the almost same as the case of the one where CNT dipping was applied on cotton fabric with MnO_2 above. They ascribed its outstanding rate capability and mass loading of the active material to large surface area within the sponge and its continuous structure that is junction-less. Nevertheless, the sponge has limitation in the perspective of volumetric energy density.

2.2.2 Conductive substrates

Hsu *et al.* [46] grew up CNTs on the CC via microwave plasma-enhanced chemical vapor deposition (MPECVD). It is

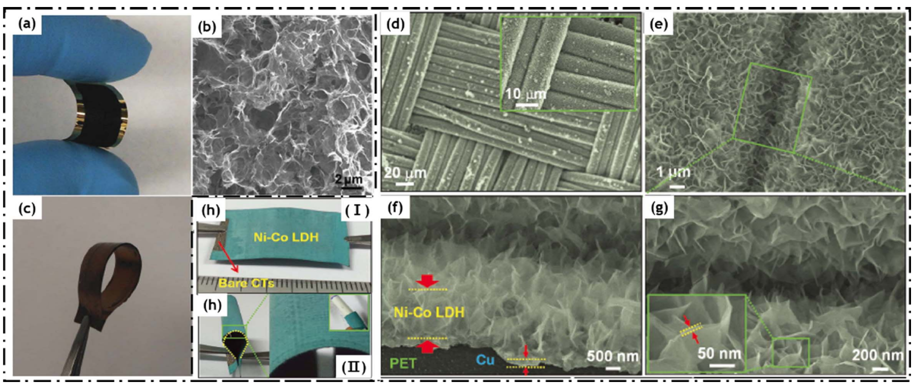

Fig. 4. (a) A functionalized graphene hydrogel (FGH) in the form of thin film, (b) SEM image of microstructures of the FGH, (c) a supercapacitor with the FGH from ref [41], (d-h) SEM images of the nickel-cobalt layered double hydroxide nanosheets on the textile substrate from ref [42]

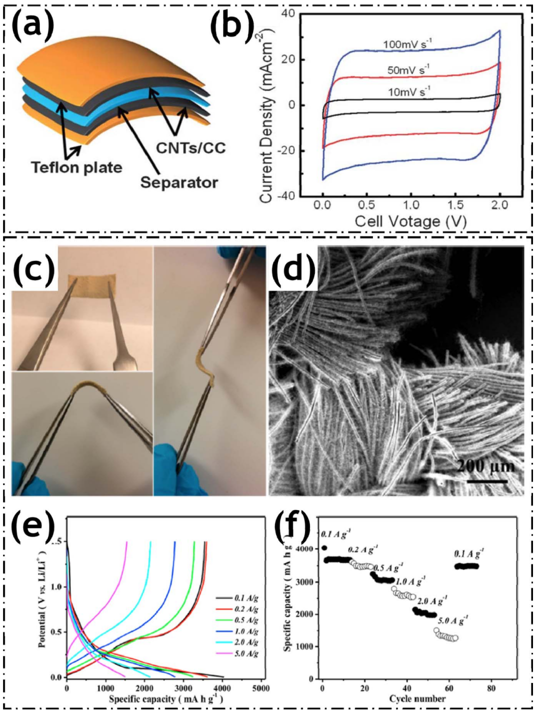

Fig. 5. (a) Schematic of CNT SC, (b) cyclic voltammetry of CNT SC from ref [46], (c) carbon-coated silicon NWs on the CF (c-Si NWs/CF), (d) SEM image of the c-Si NWs/CF, (e) charge-discharge of the c-Si NWs/CF, (f) rate capability with respect to cycle number from ref [47]

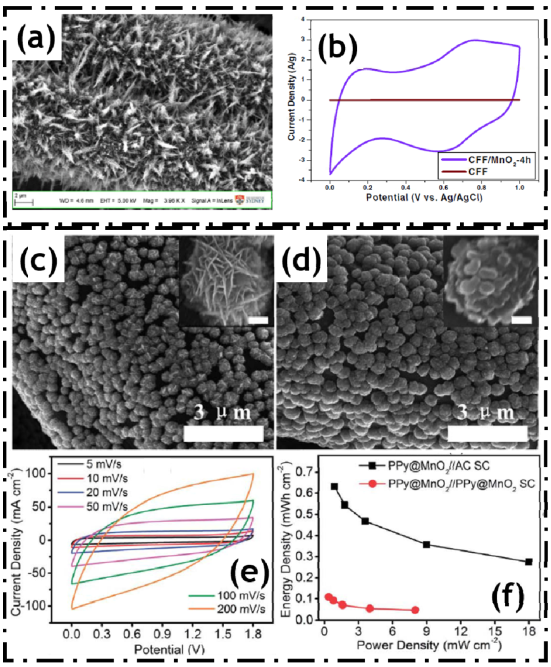

Fig. 6. (a) SEM image of Coral structure of MnO_2 on the CF (MnO_2/CF), (b) cyclic voltammetry of MnO_2/CF from ref [51], (c) SEM image of MnO_2 nanoflower on the CC (MNF/CC), (d) PPy-wraped MNF/CC, (e) cyclic voltammetry of the ASC, (f) ragon plot of the ASC from ref [52]

known that the active surface area of the electrode, which is conductive so that charge can move close to the surface for the electrical double layer or faradic reactions, is an important factor for the EDLC and the pseudocapacitance. For this reason, growing CNT on the CC is good approach in aspects of interfacial area and conductivity. They constructed a symmetric capacitor with the CNTs on the CC and 0.5 M Na_2SO_4 electrolyte (Fig. 5a). Because carbon materials almost manifest EDLC, we can see the rectangle-like cyclic voltammetry (CV) profiles like Fig. 5b. And, as scan rate increases, the crude shape does not change implying that it possesses rate-capability. It carried capacitance of 207 F g^{-1}, and energy density of 27.8 Wh kg^{-1}. Wang *et al.* [47] reported a flexible anode, which mainly consist of silicon nanowires (NWs) on the CC, synthesized by electrodeposition for seeding followed by chemical vapor deposition for the purpose of LIBs (Fig. 5c, d). Silicon is an intriguing anode material due to its high theoretical specific capacity despite large volume change it undergoes. 1D-structures like, especially nanowire, have a merit in alleviating stress caused during charging-discharging process. They achieved specific capacity of 3362 mAh g^{-1} after 100 cycles at current density of 100 mA g^{-1} in half-cell tests (Fig. 5e). Besides, even in the high current density, it can operate and restore its capacity stably when current density increase gradually and back to the initial value (Fig. 4f). Nano-structures directly grafted on the CC or CF are quite common. However, it is difficult to make strongly bonded ones without binders, as the name says, that bind active and conducting materials and attach them to the current collectors [48-50].

Fernando's group [51] implemented a SC based on the CF with MnO_2 (Fig. 6a) that is metal oxide to overcome drawbacks of carbon-based nanomaterials such as poor specific capacitance and low energy density. Manganese, of which dissolving is a problem in lithium ion battery, is a low-cost element. The SC showed 463 F g^{-1} at 1 A g^{-1} with good cyclic capacitance retention and cost efficiency. As shown in Fig. 6b, the area within the CV profiles evidently became wider. It seems like the SC has hybrid capacitance that EDLC and pseudocapacitance combine.

Tao *et al.* [52] construct ASC with two electrodes of PPy-coated MnO_2 on the CC (Fig. 6c) and activated carbon on the CC (Fig. 6d). Owing to two different operational potential windows of the electrodes, the fabricated cell can have a wide potential window that is favorable to increase energy density. The ASC can operate in the potential range from 0 to 1.8 V (Fig. 6e). It is obviously shown that ASC have better energy and power density compared to SC in Fig. 6f. In the volumetric performance, it carried 8.67 mWh cm^{-3} at 12.35 mW cm^{-3}. Moreover, CC substrates give the SC high mechanical stability and flexibility. Rather than mono-metal oxide, binary or trinary metal oxide generally have better conductivity [53]; it could vary drastically depending on temperature. Laifa *et al.* [54] synthesized $NiCo_2O_4$ nanowires (NWs) on carbon textile

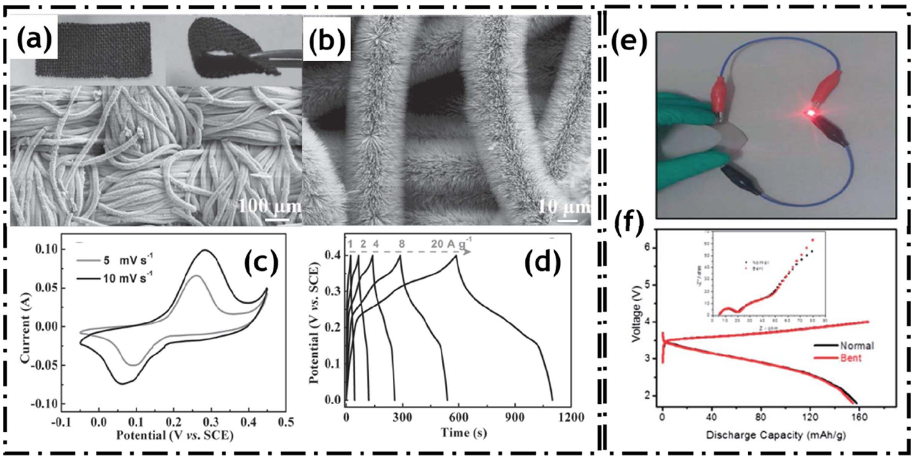

Fig. 7. (a,b) SEM image of NiCo NW on the CC (NCNW/CC), (c) cyclic voltammetry of the NCNW/CC (d) charge-discharge of the NCNW/CC from ref [54], (e) LED-on by $LiCoO_2$//TiN, (f) charge-discharge of $LiCoO_2$//TiN from ref [58]

via hydrothermal route followed by annealing process (Fig. 7a, b). The $NiCo_2O_4$ has superior conductivity to single metal oxides that consists of either the cobalt or the nickel. Besides, the $NiCo_2O_4$ NWs electrodes had especially advantages in the nature of nanostructures, of which mesoporous and 1D nanomaterial possess shortened ion diffusion path and robustness to volume change, resulting in good performance in both the lithium ion battery (LIB) and the supercapacitor (SC) [55,56]. It exhibited specific capacity of 1012 mAh g^{-1} at 0.5 A g^{-1} as a LIB electrode and around specific capacitance of around 1000 F g^{-1} as a SC electrode. As shown in Fig. 7c and d, the primary mechanism of $NiCo_2O_4$ NWs as a SC electrode, is pseudocapacitance; there is a large cathodic peak and an anodic peak. Meanwhile, transition metal nitrides have been recognized as taking candidate for their good conductivity as well [57].

Balogun *et al.* [58] demonstrated that titanium nitride nanowires based on carbon fabric (TiNN/CF) could be a good anode in LIBs. The TiNN/CF was synthesized through hydrothermal followed by thermal annealing with ammonia (NH_3). Since TiN possesses good conductivity, the anode electrode exhibited rate capability of 288 mAh g^{-1} at 1675 mA g^{-1}. In addition to this, they incorporated a flexible full cell that consists of TiNN/CF and $LiCoO_2$ (Fig. 7e, f) giving specific capacity of 157 mAh g^{-1} after 5th bending cycle. When an active material has poor in conductivity, covering the active material with conducting layer is recommendable. Li *et al.* [59] made flexible composite with polypyrrole (PPy)-coated vanadium oxide (V_2O_5) and manganese oxide on the CF. And, porous glassy fibrous paper was used as a separator after being soaked in poly vinyl alcohol (PVA) / lithium chloride (LiCl) gel. They said that PPy coating on the metal oxides enhanced conductivity of the electrodes and the cyclic stability since the PPy have a role of conducting layer as well as buffer between active material and the electrolyte concurrently. Consequently, the composite showed performance; energy density is of 28.6 Wh kg^{-1} at power density of 200 W kg^{-1}. Zhou *et al.* [60] made a flexible ASC with polyindole (pIn)-decorated V_2O_5 on the activated CC (ACC) (Fig. 8a) and reduced graphene oxide on the ACC. As like the role of PPy, pIn also give electrodes not only improved conductivity but also protecting layer as well. It facilitates superior capacitance retention of pIn-decorated V_2O_5 to non-decorated V_2O_5 (Fig. 8d). The ASC possess energy density of 38.7 Wh kg^{-1} at 900 W kg^{-1}. By cyclic voltammetry test, they also revealed that the flexible cell carries stably capacitive behaviors at different bending angles. When some devices jump into industrial practical fields beyond lab-scale, cost-effectiveness is one of considerable factors. Using inexpensive materials at the least cost of other electrochemical performance is favorable. Yang *et al.* [61] construct the ASC with low-cost metal oxides (MnO_2 and Fe_2O_3). MnO_2 nanowires were grown on the CF directly by hydrothermal method. Fe_2O_3 nanowires were synthesized via exchange metal ions from ZnO nanowires. The nanowires were grown radially on

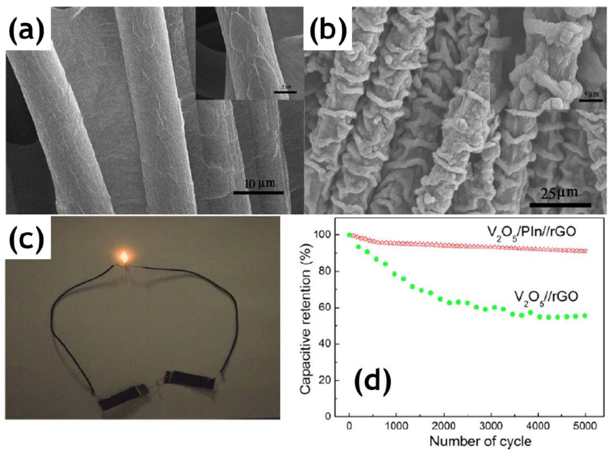

Fig. 8. (a) SEM image of reduced graphene oxide on the activated CC, (b) vanadium oxide on the activated CC, (c) LED-on by V_2O_5//Pin//rGO, (d) cyclic performance from ref [60]

the surface of carbon fiber making active materials react in wide interfacial area. Furthermore, MnO_2 and Fe_2O_3 have different potential window to react suitably. This facilitate that the ASC operate in large potential window if the electrolyte is available; the higher potential the cell operates in, the better energy density it has. And, the ASC can be implemented at the low cost relative to other FESDs.

3. GEL OR SOLID-STATE POLYMER ELECTROLYTE

Most of dangers in LIBs generally come from liquid electrolyte that is flammable, volatile, and vulnerable to leakage. In the aspects of flexible batteries, safety is the most considerable issue in it. And, non-liquid type electrolyte have been considered as an definite solution for this unsafety of LIBs. Polyethylene oxide (PEO) have been widely studied for ionic conductive polymer. The ionic conductivity of PEO itself (10^{-7} S cm^{-1}) is not enough to be applied in battery applications. For this reason, sorts of plasticizers are used to increase the conductivity at the cost of mechanical strength. A number of PEO-based polymer electrolytes incorporated with lithium salts and plasticizers have been suggested. Balo *et al.* [62] made gel polymer electrolyte (GPE) where ionic liquid (1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide (EMI-TFSI) and lithium bis(trifluoromethylsulfonyl)imide (LiTFSI) were incorporated in PEO-based polymer. And, they achieved the ionic conductivity of 2.08×10^{-4} S cm^{-1} at room temperature. Zhong *et al.* [63] make a comparison with the performance of gel electrolytes, which are synthesized with PEO, benzophenone, and several ionic liquids (IL) via UV irradiation, varying kinds of IL. As expected, capacitance increased when more IL was added into the composition. They made flexible capacitor with the one of those electrolyte and achieved 30.13 Wh kg^{-1} and 874.8 W kg^{-1} at 1 A g^{-1} with potential window of 3.5 V. Despite enhancement in conductivity of the electrolyte by IL, a trade-off in phase is not avoidable. The more IL it contains, the more liquid-likely phase it has.

Porcarelli *et al.* [64] synthesized all-solid-state polymer electrolyte (SPE) with PEO, LiTFSI, and tetraglyme using UV irradiation. They used 4-methyl benzophenone (MBP) to generate free radicals in the polymer system (Fig. 9a). And, the PEO took part of polymer matrix to maintain structural network containing the lithium salt. Additionally, the tetraglyme improves the mobility of polymeric chain as a plasticizer. The SPE are homogeneously linked and does not have voids or pores (Fig. 9b, c). It manifested conductivity more than 10^{-3} S cm^{-1} at ambient temperature retaining the phase as soft solid. Kim *et al.* [65,66] synthesized solid composite polymer electrolyte (SCPE) with ethoxylated trimethylolpropane triacrylate (ETPTA) and Al_2O_3 nanoparticles. The addition of Al_2O_3 nanoparticles have effects on suppressing growth of lithium dendrite, which is main problem when using lithium foil as anode, and favorable to distribute stress improving mechanical toughness not deteriorating formation of ionic channels that ion can move in. However, the SCPE was too brittle to get through a number of bending cycles. Polymer electrolyte is the one of the toughest part to construct FESDs. And, the ionic conductivity is usually extremely dependent to ambient temperature, to achieve both ionic conductivity more than 10^{-3} S cm^{-1} at room temperature and solid phase simultaneously still remains a challenge.

4. CONCLUSIONS

To make all-in-one electrodes, in which current collectors itself can be served as electrodes with their conductive frame, would be ideal. However, in unpredictable harsh conditions like bending cycles beyond its capability and electrochemical reaction accompanying with volume change of electrodes, its mechanical stability should not be easily guaranteed. The active materials on the flexible substrates would be more stable even though it is at the cost of energy density. Most of electrodes synthesized in this frame are in the forms of binder-free nanostructures, which have positive effects on the electrochemical performance by its own nature. The porous structure for enhancing EDLC and transition metal oxide for pseudo-

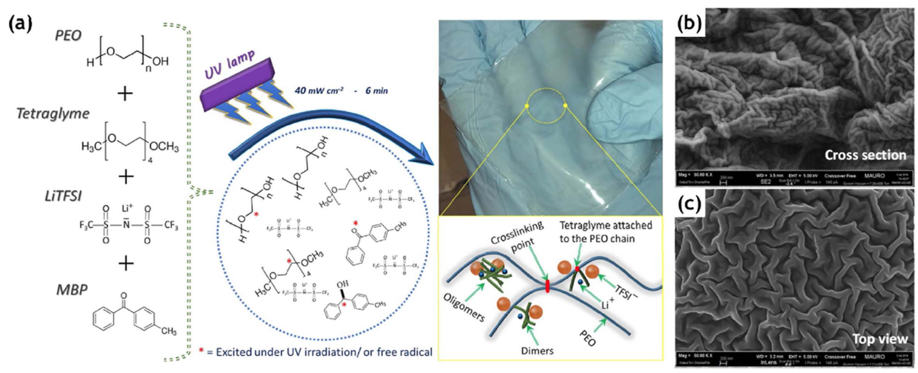

Fig. 9. (a) Schematics of preparation of interlinked solid polymer electrolyte (ISPE) and illustration (b, c) SEM images of ISPE from ref [64]

capacitance are methods to reinforce energy density. Many studies have focused on the way to increase both energy and power densities minimizing trade-off between them. On the other hand, even though mainstream in FESDs flows from batteries to SCs, ion batteries cannot be neglected from the point of view of that energy density has much priority in some specific applications such as lights-on, sensory and networking system. The FEs for ion-batteries, such as lithium, sodium, and potassium-based ones, need to be studied further in the level of full-cell. On the other hand, CF and CC-based electrodes have been considered that they have enough conductivity to role as current collector without any doubts. Nevertheless, It need be reconsidered compared to copper and aluminum foil that are conventionally available. And, non-liquid electrolytes are highly required to satisfy safety criteria for FESDs. Although many polymer gel and solid-state electrolyte have been proposed for LIBs and SCs, cost-effectiveness and ionic conductivity in the room temperature or cold situation remain as problems to overcome. There still exists critical problems in ESD of especially CF and CC-based one. However, they have large potential, which is to extend their application domain to the other's territory such as load-bearing batteries and energy-harvesting composite, using merits of carbon composite that is light, conductive, chemically stable, strong with respect to weight and so on.

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