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by Shella Santoso

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32**Fine Chemical Engineering http://ojs.wiserpub.com/index.php/FCE/ Review Trends** on **the Development of Hybrid**

Supercapacitor Electrodes from the Combination of Graphene and Polyaniline Shella Permatasari Santoso1,2* , Harry Kasuma (Kiwi) Aliwarga3, Livy Laysandra2, Artik Elisa Angkawijaya4, Felycia Edi Soetaredjo1,2, Jindrayani Nyoo Putro1, Maria Yuliana1, Felix Pasila5, Kuan- Chen Cheng6,7,8,9, Hsien-Yi Hsu10,11,12, Suryadi Ismadji1,2 1Department of Chemical Engineering, Faculty of

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Received: 2 August 2021; **Revised**: 8 December 2021; **Accepted**: 27 December 2021

Abstract

: The high

4**demand for** efficient **energy devices** leads **to the**

rapid development of

2**energy storage systems with excellent** electrochemical properties, **such as** long life cycles, **high**

cycling stability, and high power density. SC is postulated as a potential candidate to fulfill this demand. The combination of graphene and polyaniline can create SC electrodes with excellent

2**electrical conductivity, high** specific **surface area, and** high capacitance. The **graphene**

/ polyaniline hybrid electrodes represent an attractive means to overcome the major drawbacks of graphene or polyaniline non-hybrid (single) electrode materials. In this review article, the trend in the development of various graphene/ polyaniline hybrid electrodes is summarized, which includes the zero-dimension graphene-quantum-dots/polyaniline hybrid, one-dimension graphene/polyaniline hybrid, two-dimension graphene/polyaniline hybrid, and three-dimension hydrogel-shaped graphene/polyaniline hybrid. Several strategies and approaches to enhance the capacitance value and cycling stability of graphene/polyaniline hybrid electrodes are discussed in this review article, such as the addition of transition metal oxides and metal-organic frameworks, and modification of graphene into functionalized-graphene. The performance of the electrodes prepared from the combination of graphene with other conducting polymers (i.e., polypyrrole,

polythiophene, and polythiophene-derivatives) is also discussed. Keywords: hybrid electrode, nanocomposite, graphene polyaniline, conducting polymer, asymmetric supercapacitor Copyright ©2022

3**Shella Permatasari Santoso, et al. DOI: https://doi.org/10.37256/fce**.3120221077 **This is an open-access article distributed under a CC BY license (Creative Commons Attribution 4.0 International License) https://creativecommons.org/licenses/by/4.0**

/ Graphical abstract 1D fiber 0D quantum dots Graphene-based/PANI hybrid electrode 2D planar 3D foam Nomenclature Abbreviation Definition 0D zero-dimension 1D one-dimension 2D two-dimension 3D three-dimension AAm acrylamide AEESC active electrolyte-enhanced supercapacitor ANI aniline APS ammonium persulfate ATRGO amino triazine functionalized-reduced graphene oxide BTC 1,3,5-benzene tricarboxylic BET Brunauer-Emmett-Teller BDC 1,4-benzene dicarboxylic CC carbon cloth CFC carbon fiber cloth CNT carbon nano tube CP conducting polymer CSA camphorsulfonic acid CT cotton CV cyclic voltammogram CVD chemical vapor deposition EDLS electrical double-layer supercapacitor EMITFSI/PVDF-HFP ionic liquid/poly (vinylidenefluoride-co-hexafluoropropylene) ESD energy storage device F-GnO fluoro-functionalized graphene oxide FS Faradaic supercapacitor Gn or G graphene GCE glassy carbon electrode G-PC graphene-grafted PANI copolymer GFP graphene/Fe2O3/PANI GH graphene hydrogel GO GNS GP G/PANI GNS GOF GQD Gt HS LbL LiB MOF MWCNT NH2-Gn NH2-GNS NSA NW OMC PAAm PAG PAN PANI PCBM PDMS PEDOT PET PMMA PPDA PPy PS PTA PTh PVA PVDF RFC RGO RGOH SC SEM S-Gn S,N:GQD SS SSA TCTA TEM Th THF TMO TPA XPS graphene oxide graphene nanosheet graphene paper graphene/PANI graphene nanosheet fluoro-functionalized graphene oxide graphene quantum dot graphite hollow sphere layer-by-layer lithium battery metal-organic framework multi-walled carbon nano tube aminated graphene amine-functionalized graphene nanosheets 1,5-naphthalene sulfonic acid nanowhiskers ordered-mesoporous-carbon polyacrylamide p-aminophenol functionalized-graphene polyacrylonitrile polyaniline phenyl-C60-butyric acid methyl ester polydimethylsiloxane poly (3,4-ethylenedioxythiophene) polyester fabric poly (methyl methacrylate) p-phenylenediamine polypyrrole polystyrene p-benzene dicarboxylic acid polythiophene polyvinyl alcohol polyvinylidene difluoride resorcinol-formaldehyde carbon

2**reduced graphene oxide reduced graphene oxide** hydrogel supercapacitor **scanning electron microscopy**

sulfonated graphene sulfur, nitrogen-functionalized graphene quantum dot solid state specific surface area 2,4,6-trichloro-[1,3,5] triazine transmission electron microscopy thiophene tetrahydrofuran transition metal oxide triphenylamine X-ray photoelectron spectroscopy XRD X-ray diffraction 1. Introduction SCs, also referred to as ultracapacitors, are a type of ESD with

22**high power density, fast charge-discharge rate, and** long life-cycle.**1-3** SCs **are**

constructed from a sandwich of

4**two electrodes** and **are separated by a** porous **membrane** imbibed by the electrolyte in a solid, liquid, or quasi-solid state.4-7 SCs are considered safer than batteries, owing to their lower heating rate, better mechanical properties, and higher stability.8 The lightweight and flexible structure of SC also adds to its appeal as an ESD.9 SCs are widely used as regenerative-braking systems in hybrid cars and electric vehicles, energy storage, and power delivery for wearable or portable electronic devices.10-11 Despite their remarkable abilities, the relatively low energy density of SCs remains a persistent challenge in the field and their development. SCs

2**can be** classified **into two categories based on the energy storage**

mechanisms, FS and EDLS.2,12 The charging mechanism of an EDLS is based on the reversible electrostatic attraction between the charged surface of electrodes and the electrolyte ions. In this regard, electrode materials with high SSA and controlled-pore sizes are required to establish EDLSs with high energy storage capacity and specific capacitance values, wherein hierarchical-porous activated carbons are generally employed.13-14 Alternatively, FS stores charges based on the fast and reversible Faradaic (redox) reactions on the electrode surface.15 Combining transition metals and CPs is one of the most commonly used approaches to construct working electrode materials for FSs.16 However, FSs often suffer from poor electrical conductivity than EDLSs since Faradaic reactions are generally slower than the non-Faradaic process, which eventually leads to poor cycling stability.12,15 The combination of carbon-based materials and CPs in preparing the hybrid SC electrodes creates an asymmetric behavior, leading to high cycling stability and energy storage capacity.17 The electrochemical

31**performance of** SCs **is** strongly **dependent on the electrode materials**

.18 An ideal SC electrode should possess the following criteria: high SSA, controlled porosity, high electrical conductivity, high thermal and chemical stability, environmental friendliness, and low cost.18-19 Gn, a carbon-based material with a honeycomb monolayer structure, represents a promising candidate as the electrode material for SCs.20-21 Gn consists of all-sp2- hybridized carbon, contributing to its electron delocalization and high thermal and electrical conductivity. Gn is also known for its other superior properties, which includes its lightweight, high mechanical strength (~1 TPa), good physical and chemical stability,

1**and high surface area** (~2,630 **m2/g**).21-23 **The**

fresh-single layer of Gn is known to have

1**a high** theoretical **specific capacitance of** ~21 uF/**cm-2**

.12 However, it is still a major challenge to achieve such the maximum capacitance of Gn since its layers tend to encounter severe agglomeration during their preparation and utilization; thus, the expected capacitance value is far from being achieved.12,24 The addition of CPs can prevent the agglomeration of Gn (and vice versa), especially during the preparation step. CPs themselves have been widely utilized in the preparation of ESDs. Among the CPs, PANI

2**has received** considerable **attention due to its** good electrical properties **and thermal** stability that contribute to high capacitance. PANI can produce electrode materials with high specific capacitance values, often higher than EDLSs; however, the resulting SCs exhibit poor cycling stability and low mechanical stability. Therefore, the hybridization of Gn and PANI, to result in G/PANI, has been actively pursued to construct electrode materials to overcome the major drawback of using PANI or Gn alone. In this context, the G/PANI hybrid electrodes can achieve 2-5 times higher specific capacitance values than those of corresponding individual components. Furthermore, the G/PANI hybrid electrodes possess increased electrochemical active sites, facilitating the electrochemical reactions in SCs.25 The performance of several G/PANI-based electrodes is given in Table 1. Although G/PANI hybrid electrodes have been extensively developed with potential commercial applications, research is still widely conducted to enhance electrochemical properties. This review summarizes the trend in the development of G/PANI hybrid electrodes and reviews the strategies to improve the electrocapacitive properties of G/ PANI hybrid electrodes. This review begins by discussing the performance of various G/PANI electrodes, followed by the developed fabrication methods of enhancing the electrode capacitance. Next, several strategies to improve the G/ PANI capacitance are discussed, including adding functional groups to Gn and incorporating TMOs and MOFs. The hybrid electrodes based on the combination of Gn and other CPs are summarized to compare their electrocapacitive performances with G/PANI. Table 1. Electrocapacitive energy storage properties of Gn/CP-based hybrid electrodes Material Electrolyte Discharge Specific Capacitance Retention % Rate (F/g) (Cycle) U (Wh/kg) S (W/kg) Ref. GQD/PANI G/PANI G/PANI G/PANI G/PANI

31**G/PANI G/PANI G/PANI**@Fabric **G/PANI**

@Cloth G/PANI@CC Holey-G/PANI/G GO/PANI GO/PANI/paper fiber RGO/PANI RGO/PANI RGO/PANI RGO/PANI RGO-HT/PANI RGOHG/PANI RGO-Foam/PANI RGO/PANI/RFC G/PANI hybrid

14**0.5 M H2SO4 1 A/g**

~1,044 EMITFSI/PVDF-HFP* 0.22

5**mA/cm2** 87.**8 mF/cm2 0**.5 M H2SO4 **0**.2 mA/**cm2**

176 mF/cm2

14**2 M H2SO4 0.1 A/g**

500 1

10**M KOH 0.1 A/g**

578.8

10**1 M H2SO4 1 A/g**

763

101 **M H2SO4 1 A/g**

789.9 1 M H2SO4 1 A/cm2 790 F/cm2

141 **M H2SO4 5 mV/s**

246 mF/cm2

331 **M H2SO4 0.5 A/g** 793 **1**

M H2SO4 1 A/g 437 1 M H2SO4 1 mA 429

14**0.5 M H2SO4 1 A/g**

937 1 M H2SO4 2 mV/s 159.62

141 **M H2SO4 1 A/g**

432

141 **M H2SO4 0.5 A/g**

438.8 PVA/H2SO4* 0.5 A/g 424.4

331 **M H2SO4 0.2 A/g**

420 AEESC 2.6 A/g 1,120

101 **M H2SO4 1 A/g**

790 6 M KOH 1 A/g 523 ~80.1 (3,000) 10,000 74.8 (500) 82 (1,000) 90 (5,000) 88.9 (5,000) 98 (3,800) 81 (10,000) 84 (5,000) 1,000 74.5 (2,000) 82.02 (1,000) 85 (10,000) 76.5 (2,000) >80 (2,500) 80 (6,000) 91.4 (4,000) 80 (5,000) 87.6 (5,000) 117.45 12.2 μW h/cm2 17.1 μW h/cm2 448.8 226.4 μW/cm2 0.25 mW/cm2 32.2 793.3 28.21 μW h/cm2 0.12 mW/cm2 9.7 μW h/cm2 840.9 μW/cm2 11.38 199.8 55 900 10.12 456 0.31 1,178.78 25 681 9.3 11,600 17.6 98 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 10 43 44 45 S,N:GQD/PANI@Ni-Foam GO/PANI/eCFC RGO/PANI/eCFC N-doped RGO/PANI

2**2 M** KOH **1 M** H2SO4 **1 M** H2SO4 1 **M**

H2SO4 Functionalized-G/PANI hybrid 2-10 A/g 2,524-1,699 1

8**A/g** 940 **1 A/g 1**,145 0.1 **A/g**

590 100 (1,000) 84 (5,000) 100 (200) 71 450 25 92,200 46 47 47 48 G/CoFe2O4/PANI G/Fe2O3/PANI GO/Mn3O4/PANI RGO/Fe2O3/PANI@CC RGO/MnO2/PANI/Cotton RGO/RuO2/PANI RGO/NiPbTiO3/PANI RGO/LaMnO3/PANI 1 M KOH 1 M KOH

7**1 M H2SO4 1 M H2SO4** 1 **M**

H2SO4 1 M H2SO4 PVA-KOH* G/PANI hybrid with the addition of TMOs 0.1 A/g 716.4 638 96 (5,000) 92 (5,000) 0.3

15**A/g 0**.25 **A/g** 15 **A/g 2**

mV/s 0.5 A/g 2.5 A/g 829 1,124 888 723.09 1,180 111 94 (1,800) 82 (10,000) 70 (3,000) 85.14 (1,000) ~91 (10,000) 117 (100,000) 79.7 17 121 14.4 1.29 40 35 178.2 351 891 58 215.35 18,000 30 49 25 50 51 39 52 53 GO/PPy GO/PPy@MWCNT RGO/PPy RGO/PPy@CNT PPy/RGO/PPy@Cu:Ni PET RGO-hydrogel/PPy Carboxyl-G/PPy@CNT G/PPy/MnOx@Cu(OH)2 Co9S8/PEDOT:PSS/RGO S-doped GO/PTh RGO/PTh/AgNPs RGO/PEDOT/PANI PAN/PANI/GQD *Solid-state electrolyte 1 M KCl 1 M Na2SO4 PVA-H2SO4* PVA-H2SO4* 1 M Na2SO4

2**1 M** KNO3 **1 M** KCl 6 **M** KOH **2 M** KOH 2 **M** KOH **1 M** H2SO4 1 **M**

H2SO4 0.1 M H2SO4 Gn hybrid with other CPs 1 A/g 100 mV/s 1 A/g 10 A/g 2 mA/cm2 3 A/g 0.5 mA/cm2

16**10 mV/s 1** A/g **0**.3 **A/g**

4

16**mV/s 1** A/g **0**.67 **A/g**

271 64 (>2,000) 358.69 88.69 (2,000) 454 65 (10,000) 443 94.3 (10,000) 684 mF/cm2 4,000 340 87.4 (10,000) 196 mF/cm2 98.1 (5,000) 370 1,000 788.9 100 (10,000) 296 91.86 (4,000) 953.13 91.99 (1,000) 535 99 (10,000) 589.3 40.45 48.3 7 95 µW h/cm2 46.9 10.9 µW h/cm2 12/85 19.6 148 28.8 26.89 441 750 8.2 mW/cm2 1 mW/cm2 2,400 8.1 mW/cm2 6610 400.9 41.6 2,840 800 54 55 56 57 58 59 60 61 62 63 64 65 66 2. G/PANI-based hybrid electrodes 2.1 0D GQD/PANI electrodes GQDs are a 0D form of Gn with a particle size of fewer than 10 nm, which possesses several O-containing functional groups such as -COOH and -OH.67 GQDs are produced through physical or chemical exfoliation of carbon materials (e.g., fullerenes, Gt, Gn, etc.); wherein, a method such as oxidation, hydrothermal treatment, CVD, laser ablation, or plasma treatment is often employed.68-69 The usage of GQDs alone as the electrode materials has been reported to produce SC with a

26**specific capacitance ranging from** 6 **F/g to** 296.7 **F/g**

.67,70 Hybridization of PANI with GQDs can result in an SC electrode with enhanced specific capacitance. The presence of GQDs, in an adequate concentration, allows the homogeneous polymerization of ANI into PANI and uniform distribution of the particle size.68-71 As an example of GQD/PANI hybridization, Arthisree and Madhuri reported that the hybridization of PANI/GQD with PAN results in PAN/PANI@GQD, and this hybrid electrode can achieve a

7**specific capacitance** value **of** 587 **F/g at a current density of** 670 mA/**g in** 0.1 **M H2SO4** electrolyte.66 **The**

8**highest specific capacitance of the** GQD/**PANI** hybrid **was**

reported by Mondal et al.;26 wherein, the electrochemical characterizations

23**showed a high specific capacitance of 1044 F/g at 1 A/g**

and 0.5 M H2SO4. This group also showed that the addition of GQDs could control the agglomeration of the PANI particles. As revealed by the TEM characterization, agglomeration of PANI particles was observed in the electrode materials prepared without GQDs; the formation of homogeneous PANI nanofibers can be achieved with the addition of GQDs (5-15 wt%). The retardment of PANI polymerization was encountered at a higher number of GQDs addition (i.e., 20 wt%). The highest specific capacitance (1,044 F/g) can be achieved at 10 wt% of GQDs addition, while lower capacitance values (205.7-776.3 F/g) were achieved at other loading amounts of GQDs. The combination of fullerene and PANI has also been reported to produce SC electrodes with high specific capacitances.72-74 For instance, outstanding SCs

11**with a specific capacitance of** 2201 **F/g (at a current density of** 2 **A/g**

) have been reported by Ramadan et al. by integrating fullerene with PANI.73 The hybrid electrode also exhibits superior

4**cycling stability up to 1,000 cycles**

while maintaining the capacity retention at 96%. In their study, the fullerene/PANI electrode was prepared by mixing 5 mg of PCBM (a fullerene derivative) and 200 μL ANI in 0.5 M H2SO4 solution, followed by the addition of ammonium persulfate to polymerize ANI into PANI. They further observed that higher loading amounts of PCBM in the hybrid electrodes led to the formation of large-size PCBM and consequently a decrease in specific capacitance. It also reported that the specific capacitance was lower at high PCBM addition of 10 mg (1,313

1**F/g, at 2 A/g) and**

at low PCBM addition of 2.5 mg (1,582

35**F/g, at 2 A/g). A** considerable decrease **of** specific capacitance at high PCBM addition is due to the formation of large-size PCBM, leading to the reduction of surface area. Utilizing biomass as a renewable carbon source represents an attractive means to reduce raw material costs in preparing a product. For instance, the utilization of carrot juice as the carbon source in preparing carbon dots has been reported by Oskueyan et al.75 The carbon dots from carrot juice were mixed with PANI, PPy, and Gn to prepare the composite electrodes. Although biomass-derived material can reduce the raw material cost, the resulting SC electrodes exhibited relatively low stability; wherein, the retention stability is reduced to 62% after only 1,000

1**cycles. The** highest **specific capacitance** that **the electrode**

composite can reach is 296

5**F/g (at a current density of 5 A/g**). Furthermore, **the** drawback **of**

using biomass-derived materials is that they may contain some organic impurities, lowering the retention stability.76 GQDs show the synergistic effects on the electrochemical performance of CP-based (e.g., PANI) electrodes by controlling the agglomeration of the CPs. Furthermore, the superior electrochemical properties of GQDs emerge due to their high SSA, good electrical conductivity, and plenty of O-containing active groups. 2.2 1D and 2D G/PANI electrodes Flexible, small, thin, and durable are the desired specifications for modern electronic devices, pushing many researches toward developing thin, flexible, and lightweight SCs to substitute conventional bulky and heavy SCs. Flexible SCs typically consist of highly flexible thin-electrodes (e.g., 1D fiber or 2D planar shapes) and are usually supported by soft substrates. In this regard, the flexibility and mechanical robustness of the flexible SCs can be achieved through strong integration between the flexible substrate and the active electrode material.77-78 The 1D fiber or yarn-shaped SC electrodes have a promising application for portable and wearable devices due to their flexibility and tiny volume.12 A flexible SC based on the G/PANI hybrid fibers obtained from wet spinning has been reported to deliver a high specific

5**capacitance of** 87.**8 mF/cm2 at a current density of 0**.22 **mA/cm2**

. This G/PANI hybrid fiber was assembled on a PDMS substrate and then coated with a solid-state electrolyte prepared from the drying of polymer gel to construct a flexible electrode. Zhang et al.27 showed that Gn to PANI mass ratio plays a crucial factor in determining the capacitance of SC electrodes. For example, G/PANI composite fibers prepared with 0.1 M ANI show a uniform distribution of PANI nanomaterials on the surface of graphene fibers, and the resulting SC has a large

5**capacitance of** 55.**8 mF/cm2 at a current density of 0**.22 **mA/cm2**

. On the contrary, the G/PANI composite fibers prepared using 0.3 M ANI resulted in the aggregation of PANI on Gn and thus caused a reduction in the capacitance to 19.3 mF/ cm2. Changing the solid-state electrolyte from PVA/H3PO4 into ionic liquid EMITFSI/PVDF-HFP can also be adapted as a strategy to increase the areal capacitance of G/PANI fiber-based electrode. The specific capacitance was increased to 87.8 mF/cm2 in the presence of an ionic liquid electrolyte. CC has been widely utilized to prepare flexible two-dimensional SC electrodes, owing to its fibrous characteristic. The preparation method of the composite SC electrodes comprising PANI, Gn, and CC commonly refers to a chemical/ electrochemical co-deposition method, as illustrated in Figure 1, and consists of several steps: (1) dipping of CC into aqueous GO dispersion, (2) reduction of GO to RGO (optional), and (3) polymerization of ANI into PANI.33,35 For example, Wen et al. utilized a cyclic voltammetry to deposit Gn and PANI nanowires on CC to fabricate G/PANI@CC electrode composite exhibiting

1**a specific capacitance of** 793 **F/g at 0.5 A/g**

.35 Lin

2**et al**. also **reported the preparation of**

solid-stated SC electrodes through electrodeposition of G/PANI on carbon woven fabric, where

2**a high capacitance of** 790 **F**/cm2 **at 1 A**

/cm2 can be achieved by SC.33 Cotton cloth (CC) GO/CC RGO/CC PANI/RGO/CC GO GO reduction Aniline polymerization GO RGO RGO PANI CC CC CC Figure 1. Preparation of flexible SC electrode composites composed of PANI/RGO growth on CC As another example of a flexible 2D SC electrode, Jin et al.34 reported a high-performance electrode material from the combination of cotton, Gn, and PANI. In this regard, the G/PANI@cotton electrode was prepared by coating cotton yarns with Gn sheets and PANI nanowire arrays. A combination of PVA/H3PO4 was introduced as the doping agent. The resulting solid-state G/PANI@cotton electrode displayed

13**excellent cycling stability up to** 3,800 **cycles**

with a loss of capacitance retention of 2%. This flexible electrode has an

29**areal-specific capacitance of** 246 **mF/cm2**

and

29**an energy** density **of 9.7** μW h/**cm2 at a power** density **of 840 μW/cm2**

. The flexible 2D electrode can also be prepared by utilizing commercial paper as the substrate

13**reported by Li et al**.28 **The**

G/PANI@paper

12**shows an areal capacitance of** 176 **mF**

/cm2

> 24**at a current density of 0.2 mA**/cm2, **which is** ~10 **times larger than that of** Gn-**paper**

. Jin et al.34 also reported a hybrid electrode using the same material combination of Gn, PANI, and paper. This group shows that the amount of PANI deposited on paper

> 24**can be** controlled **by** controlling **the polymerization time**; wherein, **the electrochemical** polymerization time **of**

900s resulted in a high PANI mass density. The prepared hybrid electrode can achieve a

> 19**capacitance of** 123 **mF/cm2 at** a **current density of 0.5** A/**cm2**

. However, the resulting electrode material exhibited a relatively low cycling stability of 500 cycles with capacitive retention of 74.8%. Another work worth mentioning is from Xiao et al.79; this group reported the synthesis of RGO/PANI@paper through the deposition of GO and PANI nanofibers on the A4 paper through the printing technique. The RGO was formed through chemical reduction of GO@ paper using HI solution at an electrical conductivity of 340 S/m. The PANI nanofibers were then deposited on the flexible RGO paper by employing electropolymerization of ANI. The resulting flexible thin RGO/PANI@paper can achieve a

> 11**specific capacitance of** 522 **F/g at 1 A/g. The** addition **of**

another RGO layer on top of RGO/PANI@paper to produce a sandwich-like RGO/PANI/RGO@paper can further

> 26**improve the specific capacitance to** 581 **F/g**

. Meanwhile, RGO@paper achieved a much lower

> 2**specific capacitance of** 55 **F/g at the** same **current density**

. The resulting RGO/ PANI/RGO@paper also demonstrated an enhanced long-term cycling stability (85% capacitive retention) compared to PANI/RGO paper (57% retention) after 10,000 charge-discharge cycles. The establishment of flexible electronic devices is closely related to the development of stretchable and wearable electrodes. An excellent flexible electrode should be able to withstand mechanical deformation without undergoing electrical failure after bending or twisting. The critical factor in developing an excellent flexible electrode with high SSA is the precise interlayer thickness and uniform distribution of PANI and/or Gn on the substrate, which can be achieved by controlling the polymerization process. 2.3 3D G/PANI electrodes Hydrogel or aerogel represents a 3D, lightweight, and hierarchically porous material with interconnected structure, beneficial for ion transport in SCs.80-82 Furthermore, hydrogel also has high SSA, which allows the loading of a high number of active materials.81 Hydrogels from CPs possess high conductivity and excellent stretch-ability, which enables them to withstand structural collapse and extreme volume change during cycling.82-83 However, CPs-based hydrogels are not sufficiently tolerant of large and complex mechanical deformation. Hybridization of CPs with highly stretchable, compressible, and bendable polymer was postulated as a strategy to overcome this drawback. As an example of this strategy, high mechanical strength and super-elastic hydrogel electrode were successfully produced through cryopolymerization of anisotropic PVA/PANI. This hydrogel has a specific capacitance of 260 F/g and an energy density of 27.5 Wh/kg, and is able to fully recover after undergoing 100% stretching-compressing strain.83 Cellulose can serve as natural and low-cost 3D aerogel electrodes;81,84 the combination of cellulose/GO/PANI can result in an aerogel electrode with

21**an areal specific capacitance of** 1,218 **mF/cm2 (at a current density of 1 mA/cm2**

).81 The cellulose/GO/PANI aerogel preparation was initiated by the dissolution of cotton linter pulp in LiBr, followed by the dispersion of GO nanosheets. The PANI nanoclusters were then coated onto the cellulose/GO aerogel through in situ polymerization of ANI.81 Zhou et al.85 reported another stretchable Gn-containing hydrogel electrode, wherein PAAm was used as the copolymer of PANI. The resulting PAAm/G/PANI hydrogel shows an

19**areal capacitance of** 500.13 **mF/ cm2 (at 0.5 mA/cm2) and**

100%

7**capacitance retention after 10,000 cycles**. Meanwhile, **the** PAAm/**PANI**

hydrogel shows four times lower

19**areal capacitance of** 114.**5 mF/cm2**

. Another method for preparing hydrogel electrode is by fabricating the Gn particles into the 3D hydrogel instead of using a polymer. As an example of this approach, Liu et al.80 reported the preparation of RGO/PANI hydrogel film. The RGO hydrogel film was prepared by first treating GO with NaOH aqueous solution. The GO-containing solution was then dropped onto

25**a platform stacked** with **7 glass sheets in** an **autoclave**

reactor surrounded by a few milliliters of water. The reaction then proceeded hydrothermally

25**to obtain the RGO hydrogel film**

. Subsequently, PANI was deposited on the RGO hydrogel film through electropolymerization of ANI. The RGO/PANI hydrogel film can

18**achieve a specific capacitance of** 853.7 **F/g (at 1 A/g), and** the electrode is able to maintain 92.6% of the capacitance after undergoing 8,000 cycles. Chi et al.86 have reported the preparation of 3D hierarchical porous RGO/PANI foam. The foam electrode was prepared by the dipping and dry method, as illustrated in Figure 2a. In this context, a low-density three-dimensional metal foam (e.g., Ni- or Cu-foam) was utilized as the template. The alignment of

19**PANI nanowire arrays on the** RGO- **foam**

substrate

15**was achieved by** the **in situ polymerization** reaction. **The**

resulting composite can drastically increase the electrocapacitive properties, fulfilling

4**a specific capacitance of** 864 **F/g at** 1 **A/g with** capacitive **retention** of 85.6% **after 5,000** charge/discharge **cycles. The** preparation **of**

3D electrodes in the absence of support materials (known as free-standing electrodes) is another unique approach to preparing lightweight electrodes. Dissolvable polymers, such as PS and PMMA, were commonly used as sacrificial templates. The PS sphere was first employed as a rigid template for molding PANI-HS in such a process. PS was then removed by introducing a dissolving solvent (i.e., THF) to form PANI-HS particles. The schematic illustration showing the stepwise synthesis of PANI-HS is depicted in Figure 2b. For the GO hybridization step, the

2**negatively charged GO** nanosheets **were wrapped on the** surface of **PANI**

-HS particles and then electrochemically reduced to form PANI-HS@ERGO.

1**The specific capacitance of the** resultant composite **electrode**

was 614

5**F/g at 1 A/g with** a 90% **capacitance retention after**

500 galvanostatic charge-discharge cycles.87 PMMA is another type of dissolvable polymer that can be utilized as a sacrificial template to prepare PANI-HS.88 (a) Foam-scaffold Foam-scaffold/GO RGO-foam RGO-foam/PANI GO Scaffold removal GO-reduction GO Scaffold PANI PANI RGO (b) positive charge PANI negative charge Scaffold/GO/PANI Scaffold removal GO reduction 3D/RGO/PANI Scaffold material GO-sheet (c) PANI RGO PANI RGO PANI RGO Scaffold removal Scaffold material Scaffold(RGO/PANI)n (RGO/PANI)n (d) Scaffold material RGO/Scaffold PANI/RGO/Scaffold GO or RGO Aniline complex Polymerization RGO Aniline PANI Scaffold nanowire removal PANI/RGO Figure 2. Various techniques to prepare 3D composite electrodes (a) hydrogels or 3D foams as a template to form free-standing electrodes; (b) sacrificial polymer templates to produce hollow composite electrode particles; (c) LbL deposition of PANI and RGO on the sacrificial template; (d) ordered-mesoporous silica to create an ordered-mesoporous

carbon In another study, Luo et al.89 reported the fabrication of a novel three-dimensional G/PANI hybrid HS by using an LbL assembly technique. The

2**schematic diagram of the** LbL preparation method **is illustrated in Figure** 2c. **The**

prepared composite was utilized as the electrode for SC, yielding a

27**specific capacitance of** 381 **F/g at a current density of 4 A/g. The** resulting SC **electrode**

also exhibits a promising long-term

13**cycling stability with** a capacity **retention of** 83% **after** 1,**000 cycles**. Furthermore, **the**

authors claimed

7**that the specific capacitance of the composite**

could be tailored by changing the assembly cycle number. Combining 3D graphene with a porous structure and conducting PANI possibly still attracts many researchers in the next several years. A hybrid electrode decorated with well-ordered PANI nanowhiskers has been reported by Yan et al.90 In their study, the ordered-mesoporous silica (SBA-15) impregnated with sucrose/H_2SO_4 solution was used as a template to produce OMC via carbonization at 900 °C. The vertically aligned PANI nanowhiskers were then deposited on the OMC through an oxidative chemical polymerization of ANI. The preparation route of the PANI nanowhiskers/OMC composite is illustrated in Figure 2d. PANI nanowhiskers were first doped with CSA, resulting in an SC device with a specific capacitance of 470 F/g. 3. Strategies to enhance electrochemical properties of G/PANI hybrid electrodes 3.1 Incorporation of TMOs The incorporation of TMO particles has been actively pursued as an effective means

35**to improve the specific capacitance of the**

G/PANI hybrid electrodes. TMOs consist of transition metals from the d-block element with partially- filled d-subshell, rendering them good electrical conductivity properties.91-95 Several TMOs that have been used to enhance the specific capacitances of SCs are Fe_2O_3, Mn_3O_4, RuO_2, MnO_2, and so forth. The usage of nanosized TMOs can improve the electrochemical performance of SCs compared to their bulk counterpart, which can be ascribed to the higher surface area of nanosized TMOs.96-97 A ternary nanostructured PANI/RGO/Fe_2O_3 on CC (hereinafter referred to as PANI/RGO/Fe_2O_3@CC) composite hydrogels is reported to produce flexible SC electrode

4**with a specific capacitance of** 1124 **F/g (at 0**.25 **A/g**).50 **The** composite hydrogel electrode was

2**prepared via in situ polymerization of**

ANI using ammonium peroxydisulphate, in the presence of binary G/Fe2O3. The presence of Fe2O3 nanoparticles within the Gn layers can help in preventing the agglomeration of PANI nanoparticles, and thus increasing the

4**electrochemical performance of the electrode**. Moreover, **the**

presence of Fe2O3 nanoparticles also inhibits the restacking of the carbon sheets and subsequently improves the faradaic movement. The PANI nanoparticles themselves act as pseudocapacitance, which contributes to the improvement in the total capacitance. Further, they showed that

30**the synergistic effect of** the three **components in the**

PANI/RGO/Fe2O3@CC is perceptible

4**compared to other electrode materials** consisting **of**

G/PANI or Fe2O3/RGO. The G/PANI@CC electrode only yielded a

1**specific capacitance of** 793 **F/g at 0.5 A/g in** 1 **M**

H2SO4 electrolyte,35 while the Fe2O3/RGO@CC only achieved a

1**specific capacitance of** 255 **F/g at 0.5 A/g in** 1 **M**

Na2SO4.98 The low specific capacitance of Fe2O3/RGO is also partly attributed to the electrolyte solution; for example, in a similar material of study with PANI/RGO/Fe2O3@CC, the SC electrode from G/Fe2O3/PANI (Gn was used instead of RGO) was only able to

2**achieve a specific capacitance of** 638 **F/g** using KOH **electrolyte**

.49 Yasoda et al.25 demonstrated that using Mn3O4 nanoflakes to fabricate GO/PANI could result in the high- performance SC electrode with

1**a specific capacitance of** 829 **F/g at 0.3 A/g in**

20**1 M H2SO4**. Meanwhile, only **a specific capacitance of** 335 **F/g** can be achieved without the presence of Mn3O4 in the electrode matrix. The inclusion of mechanical stirring was reported to cause a decrease

26**in specific capacitance to** 643 **F/g**. The usage **of**

rare transition metal oxides, such as RuO2, to fabricate SC electrodes is reported to produce electrodes with theoretical high specific capacitance values between 1400 and 2000 F/g. However, the major drawbacks of using RuO2 lie in its expensive cost, low abundance, and high agglomeration tendency. Although the hybridization of RuO2 with carbon-based materials can help address the agglomeration issue, the high cost of this TMO remains the main barrier for its commercial application.23 The addition of PANI into RuO2/G composites can

22**further improve the capacitance of the** resultant **electrodes**

.

2**For example**, Rakhi **et al**.99 **showed** that **the**

electrode material prepared from chemically anchored RuO2 particles onto Gn nanosheets could achieve

1**a specific capacitance of** 365 **F/g** under **a scan rate of** 20 **mV/s**. Ates **and**

Yildrim39 also reported an almost 2-fold

17**increase in the** specific **capacitance of** the **RGO**/RuO2 electrode upon **the incorporation of**

PANI. In this case, the hybrid electrode prepared from RuO2/RGO/PANI achieved a specific capacitance of 723.09 F/g at 2 mV/s and maintained 85.14% of its initial capacitance after 1,000 cycles. On the other hand, the SC electrodes fabricated from RGO/RuO2, RGO/PANI, and RuO2/PANI all displayed lower specific capacitances of 347.28 F/g, 159.62 F/g, and 40.2 F/g, respectively, while pure RGO-based electrode only reached 37.5 F/g. The usage of binary metal oxide of Co/Fe has been shown to produce hybrid electrode materials with a superior specific capacitance value. Xiong et al.30 demonstrated that a ternary combination of CoFe2O4/G/PANI results in an electrode material possessing

1**a specific capacitance of** ~1,133.3 **F/g at a scan rate of** 1 **mV/s and** 767.7 **F/g at a** current density **of** 0.1 **A/g**

. By comparison, the binary electrode comprising G/PANI only achieved ~716.4 F/g (at 1 mV/s) and 329.3 F/g (at 0.1 A/g).

7**Moreover, the ternary composite electrode** exhibits **a** superior long-term cycling performance, maintaining 96% capacitance after 5,000 cycles. Similar results were obtained by Mousa et al.,100 which reported a slightly higher

1**specific capacitance** value (**i.e**., 1,123 **F/g at 1**

mV/s) of CoFe2O4/G/PANI electrode but slightly inferior cycling stability performance (98.2% retention over 2,000 cycles). The hybridization of G/PANI electrodes with other TMOs has been extensively investigated; however, the resultant electrode materials possess much lower specific capacitance, namely 325 F/g (Fe3O4/G/PANI) and 645 F/g (NiFe2O4/G/PANI). Very recently, Shaheen et al.52 reported that the utilization of ternary TMOs of NiPbTiO3 could dramatically

27**increase the specific capacitance of the** RGO/PANI hybrid. **The**

NiPbTiO3/RGO/PANI hybrid can achieve

9**a specific capacitance of** 1,180 **F/g (at 0.5 A/g)**, while pure PANI **and**

NiPbTiO3 only afforded 800

17**F/g (at** 13 **A/g) and** 384 **F/g (at** 5 **A/g**), respectively. **The**

major drawback of using TMOs

11**in the fabrication of** the **SC electrodes**

is the poor cycling stability. The free- standing TMOs and G/PANI/TMO electrodes commonly have cycling stability between 200 and 5,000 cycles, due to the volume change of TMOs in the electrolyte solution during the electrochemical process.96 The addition of carbon- based substrates, such as CC, represents an effective means to address this issue. For instance, growing the PANI/RGO/ Fe2O3@CC substrate can result in 10,000 cycling stability. By contrast, the free-standing G/Fe2O3/PANI and Mn3O4/GO/ PANI without CC only exhibited 5,000 and 1,800 cycling stability, respectively. Although the combination of TMOs/ G/PANI can be promising for preparing

7**energy storage devices with high** specific capacitance, **the**

high cost and low abundance of several TMOs, and the possibility of metal toxicity to the environment, need to be further considered for practical applications. The utilization of MOFs for preparing SC electrodes has started to emerge. Since introduced in 1995 by Yaghi et al.,101 MOFs have gained significant interest from scientific communities in various fields. MOF possesses a

22**large specific surface area and high** porosity **as** an **electrode** material, accommodating the electrolyte ions and acting as the redox-active sites. MOF materials prepared from the combination of H3BTC, H2BDC, and PTA with various transition metals (e.g., Ni, Co, Mn, and Fe) have been widely used for preparing the SC electrode. The MOFs are usually combined

11**with carbon-based material** or CPs, and **the reported**

electrodes can reach a specific

4**capacitance up to** 1,800 **F/g** and **cycling stability up to** 10,**000 cycles**

.102 Several high specific capacitance MOF-contained electrodes have been reported.

2**For example**, Saraf **et al**.103 **reported** that **a** combination **of**

Cu-MOF/RGO/GCE can result in a hybrid electrode with a

20**specific capacitance of** 685 **F/g (in 1 M** Na2SO4 **at a current density of 1**.6 **A/g**

). Meanwhile, the Cu-MOF/GCE and RGO/GCE electrodes are only able to achieve specific capacitance of 85 F/g, 410 F/g and 685 F/g, respectively. The hybridization of CNT with Ni-MOF is reported to result in an electrode

10**with a specific capacitance of** 1,765 **F/ g (in 6 M KOH, at a** current density **of** 0.5 **A/g**

), which is higher than the Ni-MOF (1,080 F/g).104 Composite electrode from the combination of MOF with carbon-based materials is shown to have higher specific capacitance than the electrode prepared from MOF alone. The enhancement of the specific capacitance can be related to the better

1**diffusion of electrolyte ions and electron transport** in **the**

composite electrode. To date, there is still no study that shows the electrochemical performance of the combination of MOF/G/PANI, and therefore, no information can be gathered. 3.2 Surface functionalization approaches The combination of functionalized-Gn and PANI has been reported to produce SC electrodes with great specific capacitance values. In this regard, N- and S-contained functional groups, such as amine and sulfonate, are frequently used to modulate the surface chemical characteristics of Gn nanomaterial.105 The incorporation of such functional groups has been demonstrated in several studies to play a role in controlling the morphology of PANI nanostructures anchored on the Gn plane. In this section, the preparation and performance of several functionalized-Gn/PANI hybrid electrodes are summarized and discussed, along with the

11**critical factors that influence the** electrocapacitive properties **of the** resultant hybrid **electrodes**

. The usage of S,N:GQDs along with PANI (PANI/S,N:GQDs) resulted in the SC electrode with

12**an ultrahigh specific capacitance of** 2,524 **F/g at a current density of** 2 **A/g**, showing **a**

2.5-fold enhancement in the capacitance compared to that of GQDs/PANI electrode.46 In this study, the S,N:GQD was prepared via a

23**hydrothermal method using citric acid as** a **carbon** precursor **and thiourea** (CH4N2S) **as** the S,**N-source**

. The PANI/S,N:GQDs composite materials are then obtained by mixing S,N:GQDs with ANI and followed by ANI polymerization in the presence of APS. A conductive mixture composed of 80 wt% PANI/S,N:GQDs, 10 wt% carbon black, 10 wt% PVDF, 0.3 mL N -methyl-2-pyrrolidone, and 0.3 mL ethanol was then painted on 1 × 4.5 cm2 Ni-foam to prepare the SC electrode. Ramadan et al.46 also reported that the aggregation of GQDs in the composites tends to occur at high GQDs content, causing the formation of many void spaces and decreasing the electrochemical performance of the electrode. The combination of 10 mg S,N:GQDs/ PANI resulted in an SC electrode with the highest specific capacitance. Interestingly, the addition of 10 mg PANI/ S,N:GQDs resulted in an electrode with the lower surface area than pure PANI, which is 21.9 and 30.8 m2/g for PANI/ S,N:GQDs and PANI, respectively. Although pure PANI has a higher BET surface area, its specific capacitance only reached 1080 F/g, which is 2.3 times lower than PANI/S,N:GQDs. The reduction in the BET surface area of PANI/ S,N:GQDs can be attributed to the intercalation of the S,N:GQDs into the PANI matrix, which can

25**facilitate the** mobility **of** electrons **and** ions **and** thus **enhance the** electrochemical **performance of the**

electrode. In an earlier study, Liu et al.106 demonstrated a comparison study toward the specific capacitance values of SC hybrid electrodes prepared from the combination of PANI with GO, RGO, N-Gn, and S-Gn. The composite electrode materials are constructed of multilayer stacking of PANI nanorods grown on micron-sized graphene sheets. More homogeneous growth of PANI nanorods were observed when the aniline monomers were paired with S-Gn. The sulfonate groups have higher electronegative intensities than the hydroxylamine groups in GO and Gn, thus stimulating the nucleation and uniform growth of PANI nanorods. The results showed that PANI/S-Gn composites exhibit the largest BET surface of 49.0 m2/g, while other composites possess a lower BET surface area ranging from 20.2 to 36.6 m2/g. According to XPS analysis, pure PANI and PANI/S-Gn show a high total content of -N= and -NH+ (i.e., ~50%), giving rise

21**to the electrical conductivity of the electrode. The**

highest specific capacitance of 863.2 F/g at 0.2 A/g and rate capability of 67.4% (581.6

4**F/g at** 5 **A/g) can be**

achieved by using PANI/S-Gn composites. In another work, Ke et al.107 reported a PANI/ATRGO hybrid electrode with a flower-like morphology that displays an impressive

1**specific capacitance of** 1,510 **F/g at a** current density **of 1 A/g and** 89% **cycling stability after** 1,500 **cycles**

. ATRGO was prepared through modification of GO with TCTA and p-phenylenediamine. Then, 2-10 wt% of ATRGO was mixed with ANI in 1 M H2SO4 to produce PANI/ATRGO composite. The highest specific capacitance of this composite material was obtained at 5 wt% ATRGO loading, resulting in the formation of homogenous flower-like PANI nanosheets anchored on graphene. In this regard, the homogenous growth of PANI nanosheets was not able to achieve at lower ATRGO. Higher loading amounts of ATRGO than 5 wt% gave disordered stacking of PANI nanosheets on the Gn surface. Such the morphologies and non-homogenous distribution of PANI nanosheets on Gn lead to the lower

18**specific capacitance values of** 697 **and** 1,216 **F/g for**

PANI/ATRGO 2 wt% and PANI/ATRGO 10 wt%, respectively. The preparation of a GOF/PANI hybrid electrode

13**has been reported by** Y. **Li et al.**108 **The**

GOF was prepared by mixing carboxylated-GO with Selectfluor (1-chloromethyl-4-fluoro-1,4-diazoniabicyclo[2.2.2]octane, a fluorine donor chemical) and AgNO3 in water. Then, 3% of freeze-dried GOF

12**was mixed with** ANI at **a weight ratio of**

3 : 100, followed by an oxidation polymerization to produce GOF/PANI composites. The as-synthesized GOF/PANI can achieve a

12**specific capacitance of** 502 **F/g at 1**.0 **A/g and** maintain 95% **cycling stability after** 600 **cycles. The**

GOF/PANI nanocomposite is constructed of densely packed granular PANI on the GOF plane. Furthermore, agglomeration of the PANI particles was also found in some regions of the nanocomposites; the PANI particles that are not nanosized are likely to cause the reduction of the specific capacitance and poor cycling cycles. In this regard, an SC electrode material with smoother morphology and nanosized PANI particles has been demonstrated to deliver enhanced cycling stability and higher specific capacitance, as proven in the recent study by Li et al.109 The combination of PAG/PANI resulted in an SC electrode

9**with a specific capacitance of** 1,092.3 **F/g** under **a scan rate of 5 mV/s**

.109 This amine group functionalized electrode was constructed by growing the PANI nanoarrays on the PAG. In this regard, PAG

16**was prepared** from **the oxidation of graphite** through **the Hummers method**

,

2**followed by** chemical **reduction** using **hydrazine. The resulting**

Gn nanosheets were then mixed with 1,4-benzenediamine in NaNO2/ H2SO4 solution to obtain PAG. The PAG/PANI active material was then prepared by mixing 20 wt% PAG with 0.91 mL ANI and ~1.4 g NSA in 100 mL of 1 M HCl solution. Polymerization of ANI into PANI was performed by using APS for 8 h. Another hybrid composite, namely PAG/PANI-w, uses water instead of 1 M HCl solution. However, the resultant PAG/PANI-w composite only achieved a specific capacitance of 585.0 F/g, much lower than PAG/PANI (1,092.3 F/g) and pure PANI (898.5 F/g). The higher specific capacitance values of pure PANI and PAG/PANI electrodes than PAG/PANI-w can be attributed to the doping effect of PANI by HCl. A solid-state SC, namely G-PC,

9**with a** tremendous **specific capacitance of** 1,701.1 **F/g at a scan rate of 5 mV/s**

, has been reported in another study by Li et al.110 The G-PC has an

5**energy density of** 15.3 **Wh/kg (at a power density of** 50 **W/kg) and** outstanding **capacitance retention** of 94.9% **after** 5,**000 cycles**. Such **a**

remarkable specific capacitance is achieved by combining NH2-GNS, ANI, PPDA, and TPA. The NH2-GNS were prepared by using the same previously reported method.109 The resultant NH2-GNS was then mixed with ANI, PPDA, and TPA to synthesize the G-PC nanocomposite. It was shown that incorporating the amine functional group causes an increase in the C and N contents while lowering the O content of Gn; the lower O content can help to reduce the moisture of Gn. The authors also showed that the loading content of NH2-GNS significantly affects the surface morphology of the resulting nanocomposites. The PANI particles form a nanofibrous structure, resulting in a smoother surface than previously reported PAG/PANI.109-110 In this regard, a quite uniform distribution of PANI-copolymer on the GNS was obtained at GNS-NH2 weight loadings ranging from 10 to 30 wt%. Higher NH2-GNS loadings of ca. 50 wt% resulted in the formation of the disordered nanofibrous structure of PANI with some large cavities, making it not suitable for efficient diffusion of the electrolyte ions and charge transport.

30**The highest specific capacitance of** 1,701.1 **F/g was achieved with**

an NH2-GNS mass loading of 20%, whereas higher and lower loading amounts result in lower capacitance values of 1,141.9-1,242.7 F/g. The electrode materials prepared without the addition of NH2-GNS and unfunctionalized GNS exhibited an evenly low

1**specific capacitance of** 859.2 **F/g and** 882.6 **F/g**

, respectively. 3.3 Gn-based hybrid composites with other CPs 3.3.1 G/PPy nanocomposites PPy is one of the most widely used p-type CPs for preparing composite electrodes due to its high conductivity (10- 1,000 S/cm) and ease of synthesis by chemical and electrochemical methods. Similar to other PANI cases, the grafting of carbon-derived nanomaterials to PPy can

2**improve the cycling stability of the**

resultant hybrid electrodes during charge and discharge cycles.

2**For instance, Wang et al**.111 reported **a** novel technique **to** produce **a**

composite RGO/PPy porous electrode, based on the in situ galvanostatic polymerizations. The resultant composite exhibits a BET surface area of 108 m2/g and outstanding electrochemical performance, including

1**high specific capacitance** (224 **F/g at** 240 **A/ g**

), energy and power densities of 2.5 Wh/kg and 38.6 kW/kg, respectively,

13**and good cycling stability** (17% **capacitance** loss **after**

5000 charge-discharge cycles). In another study, Zhang and Zhao112 demonstrated the

15**direct coating of** PPy **on RGO sheets**

through

15**in situ polymerization process to** produce PPy/**RGO**

composite. The authors found that ethanol as a solvent can improve the

15**dispersion of RGO sheets** and subsequently promote **the diffusion and growth of** PPy over **the** RGO **surface**. The specific capacitance **of**

8**the** PPy/**RGO composite** electrode **was** 248 **F/g at** 0.3 **A**

/g with capacitance retention of 80% over 1,000 cycles of charge/discharge. The hydrothermal method has also been utilized by Gu et al.113 to prepare a 3D RGO/PPy composite hydrogel. In this study, KMnO4 was used as the oxidant in the polymerization of pyrrole monomers to form PPy on the surface of GO sheets at 95 °C for 24 h. Under this polymerization condition, GO sheets are

20**reduced and** subsequently **self- assembled** to create **a 3D porous structure**

. The SSA of the PPy/RGO composite hydrogel was 331 m2/g, yielding

2**a specific capacitance of** 356 **F/g. The** capacitance properties **of**

the as-prepared composites were influenced by the uniformity and thickness of the PPy coating on Gn. Sun et al.114 employed a spontaneous assembly or in situ redox process without an external oxidizing or reducing agent to produce 3D RGO/PPy hybrid aerogels for SC electrodes. The composite with excellent electrochemical performance (

18**specific capacitance of** 230.8 **F/g at 1 A/g, and retention of** 58.26% **after** 50 **cycles**

) and mechanical properties was obtained using their method. The

2**strong π-π interaction between** PPy **and**

RGO is responsible for the excellence of the mechanical property of the composite. A recent study by Zhang et al.59 shows a clear correlation between the specific surface area and the specific capacitance of the resulting electrode made of freestanding PPy@RGO hydrogel.

17**The specific surface area of the**

PPy@RGO hydrogel (hereinafter referred to as PPy@ RGOH) can be modulated by varying the electropolymerization time of PPy from 0 to 30 s. The PPy@RGOH obtained at an electrochemical polymerization time of 20 s (i.e., PPy@RGOH-20 s) possesses the highest specific surface area of 231.2 m2/g, while the PPy@RGOH-10 s and PPy@RGOH-30 s only exhibited 164.7 and 152.0 m2/g, respectively. The specific surface area values of PPy@RGOH composites are influenced by the formation of aggregates of the active electrode materials (e.g., RGO and PPy). A severe aggregation of these active materials can lead to the destruction of the electrode network structure, thus reducing the rate of charge transfer and decreasing the specific capacitance of the electrode. Furthermore, the aggregation of CPs is generally observed to be more at excessive loading amounts.112,115-116 3.3.2 Gn nanocomposites with PTh and their derivatives The combination of PTh and GNS as SC electrode materials has been demonstrated in several studies, although the number of publications is relatively more minor compared to those using PANI and PPy. Among various PTh derivatives, highly conductive PEDOT is of particular interest for preparing high-performance SC electrode materials.

2**For example**, Lee **et al. reported the** fabrication **of a**

thin multilayer G/PEDOT film

4**using an in situ electrochemical** deposition **method**

.117 SEM analysis proves the successful deposition of the G/PEDOT multilayers.

8**The specific capacitance** value **of the composite** thin **film**

with six Gn layers was found to be 154 F/g, and this value decreased slightly to 132.44 F/g (86%

17**capacitance retention) after** 1,**000 charge** and **discharge cycles**. Zhang **and**

Zhao also employed PEDOT to fabricate RGO/PEDOT composite for high-performance SC electrodes. In their study, PEDOT was directly coated on RGO sheets via an in situ polymerization process.112 Ates

8**et al. demonstrated the** fabrication **of a** ternary **composite**

material from the combination of RGO, Ag, and PTh (RGO/Ag/PTh).64 The electrode with

1**a specific capacitance of** 953.13 **F/g (at** a **scan rate of** 4 **mV/s**) was achieved at **a** GO/Th ratio **of**

0.2. The prepared ternary nanocomposite also displayed

13**excellent cycling stability (91**.88%) **after** 1,**000**

charging/discharging cycles. Azimi et al.118 investigated the electrochemical properties of several PTh nanocomposites with GO and RGO. They found that incorporating GO and RGO to PTh nanoparticles could

4**enhance the electrochemical** properties **of the** resultant **electrodes**

. Specifically,

9**the specific capacitance of** GO/PTh, **RGO**/PTh, **and**

PTh

8**at a scan rate of 5 mV/s was** 16.39 **F/g**

, 28.68 F/g, and 3.5 F/g, respectively. 4. Summary and future perspective In summary, the development and breakthrough of the researches in the improvement of the electrochemical properties and cycling stability of the G/PANI hybrid electrodes have been reviewed. Incorporating Gn can facilitate the homogeneous distribution of PANI nanoparticles, thereby resulting in an SC electrode with better electrochemical properties. The synergistic effect of G/PANI comes from the π-π stacking interaction between the aromatic rings or the two compounds (Figure 3a), which boosts the electrode conductivity by facilitating the electron donor-acceptor transfer (Figure 3b). Through detailed analysis on the properties of G/PANI hybrid electrode, it can be postulated that three factors should be considered to produce electrodes with enhanced capacitance and cycling stability, including 1) composition and the ratio of Gn and PANI; 2) morphology, structure, and size of the materials; 3) hybridization with other materials. The proper amount of Gn and PANI is a crucial aspect to prevent the aggregation of the electrodes particles, and to control the morphologies and size of the particles. This effect

2**is reported in work** by Xia **et al**.49 **The**

SC electrodes prepared at a low amount of PANI, i.e., (G/Fe2O3/PANI)0.5 prepared from G/Fe2O3 to ANI

1**at the ratio of** 2:**1**, can achieve **a specific capacitance of** 638 **F/g**

(Figure 4a). Meanwhile, the specific capacitance is decreased for 1.7-3.2 times at high G/Fe2O3 to ANI at the ratio of 1:1, 1:5, and 1:10, respectively; where the resultant electrode coded as (G/Fe2O3/ PANI)1, (G/Fe2O3/PANI)5, (G/Fe2O3/PANI)10, respectively.49

1**The significant** decrease **in specific capacitance was** due to **the** occurrence **of**

aggregation, which inhibits the smooth ion transfer within the electrode matrix. Higher specific capacitance also can be achieved by combining several conducting materials. The usage of single electrode particles, such as Gn, PANI, and Fe2O3, only

1**resulted in a** low **specific capacitance of** 33-198 **F/g**

(Figure 4a). Meanwhile, combining the three materials can significantly enhance the specific capacitance to 638 F/g.49 (a) (b) NH NH π-π stacking n n e - π-π stacking OH n e - i n g N H OH n d H o n N H - b Figure 3. (a) Schematic illustration showing the bonding interaction between PANI and RGO. (b) Charge transfer process between PANI and RGO The establishment of material with nanosize, homogenous structure, and controlled morphologies is a crucial point in producing SC with excellent properties. The introduction of conductive materials (e.g., TMOs and MOFs) and/ or functionalization of the materials are approaches that can be employed as a strategy to enhance the electrochemical properties of SC. The usage of TMOs may result in higher specific capacitance value and better electron transport for the electrode; however, their high cost, low availability, and high toxicity risk are major challenges for their application. The type of TMOs also

2**plays a** vital **role in** affecting **the** electrochemical **properties of the**

resulting electrode. As shown in Figure 4b, a different value of specific capacitance was obtained by using a different type of single TMOs. The usage of binary TMOs (instead of single TMOs) can enhance the specific capacitance of the electrode. Nevertheless, many TMO combinations are available and waiting to be studied. Therefore, a complete understanding of the properties and conductivity of each TMO will help in determining the best binary combination. Other

10**strategies to enhance the** specific **capacitance of** the electrode include the incorporation of MOFs and surface functionalization of Gn. The incorporation of MOFs can assist in increasing the SSA and porosity of the electrodes, thereby facilitating the diffusion of electrolyte ions; however, the high cost and complexity of the MOFs production process is still a challenge. Surface functionalization of Gn was also postulated as one of the strategies for achieving high-performance electrodes. However, the poor cycling cycle and the tendency of particles agglomeration often hindered their industrial-scale application. While each strategy has its advantages and disadvantages, one should review the intended use of SC to determine the best electrode composition. Numerous promising methods and approaches to improve the performance of G/PANI hybrid electrodes have been demonstrated; however, there are still many challenges and research gaps awaiting to be answered. Although thousands of works related to G/PANI hybrid electrodes have been published, only hundreds of them demonstrated SC with cycling stability >10,000 cycles. While studies of SC preparations with various combinations of materials (including materials not discussed in this review) are progressing rapidly, the knowledge about the fundamental charge-discharging mechanism for these new SC combinations is still limited. Therefore,

21**it is still a challenge to** optimally **design** SCs materials **with excellent**

electrochemical properties and should be the focus of future research. (a) 800 Single Binary Ternary Specific

2**capacitance (F/g**) 700 **600 500 400 300 200 100**

0 (b) Specific capacitance (F/g) 1400 1200 1000 800 600 400 200 0 Fe2O3 I G n G/PA G/Fe2O3 N I Fe2O3/PA(NG/Fe2O3/PANI)10Fe2O3/PANI)5Fe2O3/PAN(GI)/1Fe2O3/PANI)0.5 I P A N (G / (G / Fe2O3 M n3O4 R uO2 Fe3O4 C oFe2O4 NiFe2O4 TiO3 N i P b Single TMOs Binary/Ternary TMOs Figure 4. (a)

16**Comparison of the specific capacitance of** some G/**PANI electrodes**

with various TMOs added, adapted from Ref. 49 (b)

16**Comparison of the specific capacitance of**

single, binary, and ternary hybrid electrodes comprised of Gn, PANI, and Fe2O3, adapted from Refs. 25, 30, 50, 52, 98-100 Acknowledgment This project was supported by

3**Widya Mandala** Surabaya **Catholic University through** an **internal research grant**

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34**Conflict of interest statement The authors declare that there is no conflict of interest. References**

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