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Review Article

A Review on Cutting Edge Technologies of Silicon-Based Supercapacitors

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Despite Si-based materials and their derivatives have recently emerged as potential electrode materials in advanced energy conversion and storage applications, a review article has not been reported hitherto for Si-based supercapacitors. In this review, the representative progresses of Si-based materials have been illustrated including synthesis, properties, surface modification, and electrochemical properties. A variety of nanomaterials are presented regarding the electrode material design and booming device constructions. Effective strategies for the preparation of Si-based materials and their derivatives are summarized especially including silicon/silicon carbide nanowires, silicon substrates, silicon particles, three-dimensional silicon structures, and silicon-based doping materials. Meanwhile, the overall behaviors in supercapacitor application have been illustrated in terms of specific capacitance, rate capability, cycling life, and energy density. Furthermore, large-voltage microsupercapacitors are outlined for next-generation integration devices.

1. Introduction

In past decades, the development of sustainable energy storage devices has been witnessed as one of the most important topics owing to the booming growth of renewable energy sources and devices such as electric vehicles, digital devices, and pulsing techniques [1–3]. Among the energy storage systems, supercapacitors are of significance with representative intrinsic advantages including high power density, superior rate capability, short charging time, safe operation, a long cycle life, and moderate energy density [4–6].

In general, supercapacitors can be categorized into electrochemical double-layer supercapacitors (EDLCs) and pseudocapacitors (PCs) according to the energy storage mechanism. Firstly, the energy storage mechanism of EDLCs relies on the ion of adsorption/desorption on the electrode surface-electrolyte interface. Secondly, PCs store electric charges based on rapid redox reaction at the electrode surface. Compared to PCs, EDLCs are widely used to offer high power density and superior cyclic stability but suffer from moderate energy density [7–9].

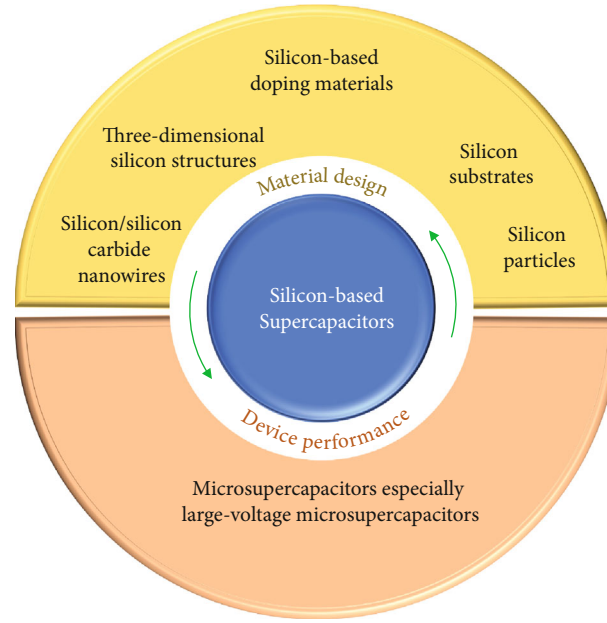
Silicon-based materials and their derivatives have attracted considerable attention as electrode materials for applications in supercapacitors with favorable performances due to the following beneficial features [10–13]. Firstly, silicon is abundant in natural sources and then silicon-based materials are featured with low price. Secondly, silicon-based materials for extended application attributed to their high stability, nontoxicity, and well-established fabrication technique. Thirdly, silicon is high carrier mobility, which renders good electrolyte accessibility and short diffusion distances for enhancing mass transport efficiency. Literatures have witnessed silicon-based materials and their derivatives with great potential in energy storage systems [14–17].

Microsupercapacitors in miniaturized, reliable, and efficient characteristics are required to meet the development of portable and integrated power sources [18–20]. When applied in an energy storage system, microsupercapacitors perform high power density, fast changing/discharging time, and cycle stability as other types of supercapacitors. Besides, microsupercapactiors can be integrated and scaled down with electronics or other energy conversion and/or storage

systems on the same chip [21–23]. Silicon as the basic semiconductor has been employed in EDLC-based microsupercapacitors.

Plenty of work has involved Si-based materials and their derivatives as electrode materials instead of EDLC electrode materials to broaden the application in portable devices [24]. Miscellaneous Si-based materials and their derivatives have attracted scientific focus owing to their superior electrical conductivity and favorable surface area.

Nevertheless, the use of Si-based materials and their derivatives as electrode materials in supercapacitors has not been reported hitherto. In this review, we have summarized the current state-of-the-art improvements in Si-based supercapacitors concerning the material nanostructure construction and booming device measurement as shown in Scheme 1. From the electrode material perspective, effective strategies were explored for the preparation of silicon nanostructures including silicon/silicon carbide nanowires, silicon substrates, silicon particles, three-dimensional silicon structures, and silicon-based doping materials. Besides, the as-prepared Si-based materials and their derivatives are constructed in microsupercapacitor devices and the device performances are revealed in terms of specific capacitance, cycling stability, rate capability, energy density, power density, and so on. Furthermore, high-voltage microsupercapacitors are illustrated with the optimized electrolyte to improve the unsatisfied energy density of supercapacitors compared to batteries.



Scheme 1: The brief illustration of silicon-based supercapacitors.

2. Material Synthesis

Generally, controlling the morphology and structure is vital to their performance in supercapacitors. In the past decade, some advanced methods have been applied to synthesize and construct Si-based electrode materials in various sizes and morphologies (nanowire, nanosheet, particles, etc.). To achieve superior electrochemical performances, the electrode materials can be modified as follows. Firstly, a high surface area is desirable to offer more accessible paths for efficient ion diffusion. Secondly, excellent electrical conductivity is a benefit for decreased internal resistance so that it facilitates the accumulation of electrostatic charges.

2.1. Si Nanowire. Silicon nanowires (SiNWs) with one-dimensional (1D) nanostructure have awakened intensive attention for supercapacitive application [25]. Compared to traditional supercapacitor materials, SiNWs displayed further advantages in microelectronic devices attributed to their compatibility with standard fabrication and integration processed for the construction of microelectronic devices.

2.1.1. Bare SiNW-Based Materials. SiNWs have been used in the self-charging power packs with a combination of hybrid solar cell and supercapacitor [26]. Figure 1(a) has illustrated the scheme of the hybrid devices wherein the top was the solar cell for harvesting energy and the bottom was the supercapacitor for storing energy. A supercapacitor in the hybrid device was based on SiNWs with the morphology of vertically aligned arrays as shown in Figure 1(b).

Chemical vapor deposition (CVD) is one of the effective strategies for the preparation of silicon nanowires (SiNWs). Thissandier et al. have obtained SiNWs grown by CVD on a silicon wafer [27]. Moreover, SiNWs can be synthesized by a one-step electroless metal-assisted etch technique [28]. Compared to the CVD method, this process the SiNWs in this method can be produced by low-temperature electrochemical etching, which avoids the metal catalyst preseeding step and high-temperature growth conditions. The SiNW arrays were produced via wet etching of Si substrates using an oxidant/etchant bath with different times under the bath temperature of 50°C, as shown in Figure 1(c). Followed by coating with a silicon carbide passivation layer, the SiC/SiNW electrode achieves capacitance of $1.7\,mF\,cm^{-2}$, robust cycling stability, and outstanding rate capability.

Unique hybrid structures based on silicon nanowires have been developed for a supercapacitor via a novel yet simple hybrid material passivation strategy [29]. The hybrid structures were prepared via the deposited $NiCo_2O_4$ (NCO) nanoflakes on the surface of SiNWs. This strategy can alleviate the rapid oxidation of SiNWs in an aqueous electrolyte, and then, the supercapacitors achieved increased energy storage capacity and aqueous electrolyte stability.

2.1.2. SiNW-Based Composite Materials. To elaborate the capacitive properties of electrodes, cost-effective and scalable approaches have been proposed. For example, poly(3,4-ethylenedioxythiophene) (PEDOT) was coated onto chemical vapor deposition- (CVD-) grown silicon nanowires (SiNWs) by electrochemical deposition [30]. Supercapacitor investigation displayed enhanced electrochemical capacitance of as-obtained PEDOT@SiNWs with a high areal capacitance up to $17\,mF\,cm^{-2}$ at $100\,mV\,s^{-1}$ under a three-electrode cell configuration.

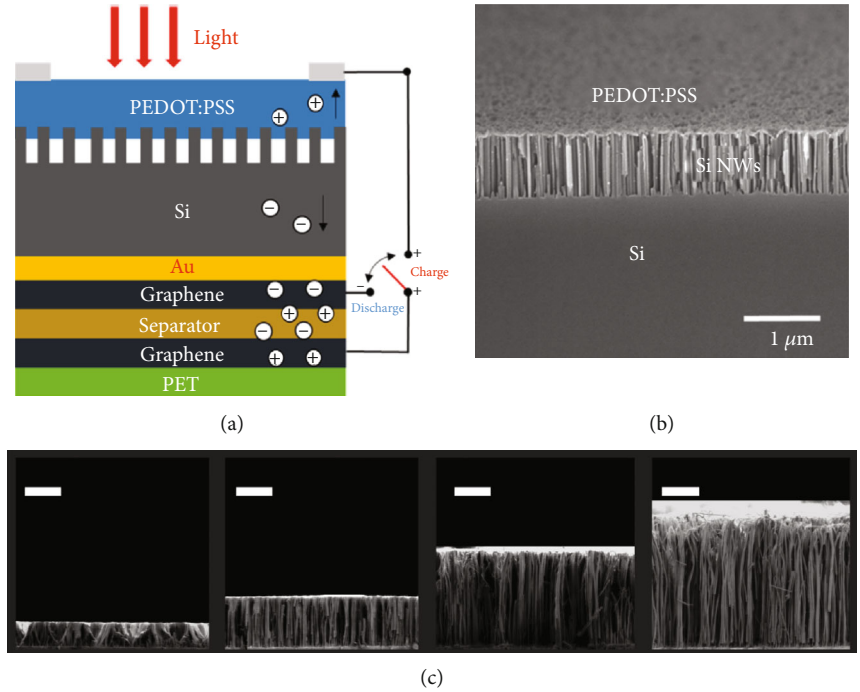


FIGURE 1: (a, b) The scheme of the self-charging power packs with a combination of hybrid solar cell and supercapacitor and the SEM images of the silicon nanowires in the hybrid devices. Reprinted with permission from Ref. [26]. Copyright 2018, American Chemical Society. (c) Etching SEM images of the hybrid composed by silicon carbide and silicon nanowire arrays for different times. Reproduced with permission from Ref. [28]. Copyright 2012, AIP Publishing.

Carbon derived from sucrose was coated over silicon nanowires by drop casting. Superior to bare SiNWs (96 μF cm^{-2}), carbon@SiNWs displayed a significantly higher areal capacitance of 3.3 mF cm^{-2} [31]. Novel architecture has been developed by coating SiNWs with graphene by an easy and efficient electrophoretic deposition technique [32]. Compared to bare SiNWs, the graphene@-SiNWs show superior capacitance with up to four times values. Manganese dioxide (MnO_2) nanoparticles have been applied for SiNWs grown at low temperature using hot-wire chemical vapor process (HWCVP) [33]. The as-prepared SiNW@MnO_2 hybrid exhibited stable specific capacitance of 2.1 mF cm^{-2} in 1 M Na_2SO_4.

SiNWs decorated with ruthenium nanoparticles (Ru/SiNWs) have been proposed through a common vapor-liquid-solid (VLS) growth mechanism [34]. Herein, a simple electroless process is used to deposit Ru nanoparticles. At first, Ru/SiNW-based microsupercapacitors were fabricated in Na_2SO_4 aqueous electrolyte and displayed favorable electrochemical performances with a specific capacitance up to 36.25 mF cm^{-2} at 1 mA cm^{-2} and high stability over 25000 cycles. Secondly, Ru/SiNWs were applied as symmetric electrodes to construct a solid-state supercapacitor in polyvinyl alcohol/sulfuric acid electrolyte. The solid-state device exhibited the specific capacitance of 18 mF cm^{-2} at 1 mA cm^{-2}, a high specific power density of 0.5 mW cm^{-2}, and outstanding stability over 10000 galvanostatic charge-discharge cycles.

The electrical conductivity of SiNWs is of significance to enhanced electrochemical performances, and enhanced electrical conductivity of SiNWs can be achieved by doping. SiNWs as a pure powder have been produced by a versatile, low-cost, and easily scalable synthesis method [35]. Air-stable diphenylsilane, gold nanoparticles, and micron-sized NaCl particles served as Si source, catalyst, and sacrificial support, respectively. The diameters of SiNWs can be controlled precisely at 10 nm with a high production yield. Phosphorus-doped SiNWs proved required high electrical conductivity. In this work, doping SiNWs have been fabricated with N-type doping by adding diphenylphosphine as a nitrogen source at various concentrations. The electrode of doping SiNWs exhibited outstanding electrochemical properties in symmetric supercapacitor devices with a specific capacitance of 0.25 mF cm^{-2} and stable retention over one million cycles.

Soam et al. have integrated an on-chip microsupercapacitor based on SiNWs by hot-wire chemical vapor process [36]. The growth of SiNWs at low temperature of 350°C in the electrode fabrication renders them prospective flexible electronic electrode materials. The on-chip microsupercapacitors have been tested in an ionic electrolyte with an areal capacity of 13 μF cm^{-2} and energy density of 0.1 μJ cm^{-2}.

SiNWs with boron-doped diamond coating have been employed as electrode materials for microsupercapacitor using an aqueous electrolyte (0.1 M $LiClO_4$) [37] and ionic liquids [38]. The nanometric boron-doped diamond coating played important roles in enhanced capacitive performances as follows: Firstly, the electrochemical window can be up to 3 V in the aqueous electrolyte and 4 V in the ionic liquids due to the deposition of the diamond coating. The enhanced voltage window rendered enhanced energy density. Secondly,

the presence of coating can decrease rapid silicon oxidation and improve the stability. Consequently, the device in the aqueous electrolyte exhibited superior capacitive performances such as high specific capacitance (0.4 mF cm^{-2}), high power density (50 mW cm^{-2}), and outstanding cycling stability over $2 * 10^6$ cycles. Besides, the properties of the device in the ionic liquids reached an areal capacity of 10^5 μF cm^{-2}, energy density of 84 μJ cm^{-2}, and good stability over 10000 cycles.

Besides, diamond-coated SiNWs deposited with nanometric poly(3,4-ethylenedioxythiophene) films were used as electrode materials [39]. The supercapacitors were measured in ionic liquid and performed a specific capacitance (140 F g^{-1}) at 1 mV s^{-1}. In addition, the as-prepared modified electrodes were fabricated into symmetric planar microsupercapacitor and investigated with an outstanding energy density of 26 mJ cm^{-2}, power density of 1.3 mW cm^{-2}, Coulombic efficiency, and stable cycling stability over 15000 cycles under the voltage of 2.5 V.

A conformal nanolayer of TiN was deposited on silicon nanorod arrays via an atomic layer deposition technology. Herein, silicon nanorod arrays were obtained through a cyclic deep reactive ion etching process and used as a scaffold [40]. The TiN-coated silicon nanorods revealed enhanced performance of 1.55 mF cm^{-2} at the scan rate of 2 mV s^{-1} and 0.95 mF cm^{-2} at the scan rate of 1000 mV s^{-1}, respectively.

MnO_x can be used to decorate carbonized porous silicon nanowires through ecofriendly and cost-effective processes, and the hybrid materials were denoted as MnO_x/C/PSiNWs [41] as shown in Figure 2(a). In brief, firstly, porous silicon nanowires were obtained from a silicon wafer by chemical etching under the assistance of metal. Secondly, porous silicon nanowires were obtained by chemical vapor deposition technique using the ultrathin graphitic carbon sheath as templates, which were denoted as C/PSiNWs. Thirdly, the above C/PSiNWs were coated by ultrathin MnO_x layer during an electroless deposition process. This electrode material system based on MnO_x decorated SiNWs exhibited excellent electrochemical behavior in EMIM-TFSI ionic liquid electrolyte. The specific capacitance reached 635 F g^{-1}, and an areal energy and power density achieved 100 mW cm^{-2} and 0.46 mWh cm^{-2}, respectively.

Hierarchical MnO_2@silicon nanowire heterostructures on a silicon wafer were fabricated with Li-ion doped 1-methyl-1-propylpyrrolidinium bis(trifluromethylsulfonyl)imide (PMPyrrBTA) ionic liquids as an electrolyte for microsupercapacitors, which have been illustrated in Figure 2(b) [42]. The as-fabricated devices performed a voltage of 2.2 V and displayed excellent capacitive behaviors with a high areal capacitance (13 mF cm^{-2}), high energy density, and excellent cycling stability. Enhancement of electrochemical capacitance of SiNWs has been achieved by modification with MnO_2 via a chemical electroless method [43]. The improved capacitance benefited from pseudocapacitive behavior of the MnO_x coating.

Besides metal oxides/hydroxides, conducting polymer is one of pseudocapacitive materials for supercapacitors such as polythiophene, polypyrrole, and polyaniline. Compared to the bare SiNWs, conducting polymer coating can induce extra pseudocapacitive behavior for improved electrochemical properties. For example, a hybrid symmetric microsupercapacitor has been developed using silicon nanowires with poly(3,4-ethylenedioxythiophene) coating as an electrode in an ionic liquid electrolyte [44]. The devices were investigated with a specific energy density of 10 W h kg^{-1} within the voltage of 1.5 V and long electrochemical stability.

CrN has been used to modify SiNW arrays using a one-step bipolar magnetron sputtering method, and the hybrid SiNW-CrN materials were applied in microsupercapacitor application in 0.5 M H_2SO_4 electrolyte [45]. To examine the factors, different thicknesses of the CrN layers were grown on SiNWs under the similar procedures. The optimized hybrid SiNW-CrN materials exhibited an areal capacitance of 180 mF cm^{-2} at 5 mV s^{-1}, which is 116 times more than that of pristine SiNWs. Porous nickel bundled silicon nanowires have been used as a supporter to disperse graphene sheets via the electrochemical exfoliation approach [46]. The direct and close contact between graphene and Ni-SiNWs resulted in enhanced conductivity and active sites of the electrode.

2.2. Si Substates. Silicon substrates have been employed in microsupercapacitor. For example, vertically oriented graphene nanosheets were deposited onto doped Si substrates via a simple electron cyclotron resonance-plasma enhanced chemical vapor deposition approach [47]. The SEM images and TEM images illustrated in Figures 3(a)–3(c) have revealed the vertically oriented graphene films on the Si substrate uniformly and densely. And Figure 3(d) concludes the function of deposition time with the thickness of graphene film on the Si substrate. The as-prepared electrodes were fabricated in symmetric microsupercapacitor devices, and aprotic ionic liquids were used as electrolytes with the voltage of 4 V. The electrochemical performances revealed the quasi-ideal capacitive behavior with the specific capacitance of 2 m cm^{-2}, the power density of 4 mW cm^{-2}, and the energy density of 4 μWh cm^{-2}.

Planar Si substrates have been used as a basic component for the growth of TiO_2 nanotube arrays [48]. After sputtering and anodic oxidation, the hybrid materials Si/TiO_2 have been obtained and Si/TiO_2@MnO_2 were obtained with the following electrochemical reduction and MnO_2 deposition. The ordered TiO_2 nanotube arrays with a high surface area were favorable for ion diffusion, and the electrochemical reduction benefited for enhanced electroconductivity for improved electron transport. The as-prepared hybrid Si/TiO_2@MnO_2 displayed the areal specific capacitance of 5.6 mF cm^{-2} at the current density of 0.05 mA cm^{-2}.

Si substrates can offer basal for the growth of metal silicide nanowires such as Co_2Si [49]. The single-crystalline Co_2Si nanowires/Si materials can be used as freestanding electrodes to construct on-chip microsupercapacitor in ionic liquid electrolytes. High performance was investigated with an areal capacitance of 983 μF cm^{-2}, energy density of 629 μJ cm^{-2}, and 94% retention after 4000 cycles. Silicon microchannel plates have been employed as substrates for the deposition of $Co(OH)_2$/graphene/Ni as the active electrode materials in microsupercapacitors [50].

The three-dimensional nano-Ni electrodes based on Si microchannel plates have been applied in electrochemical

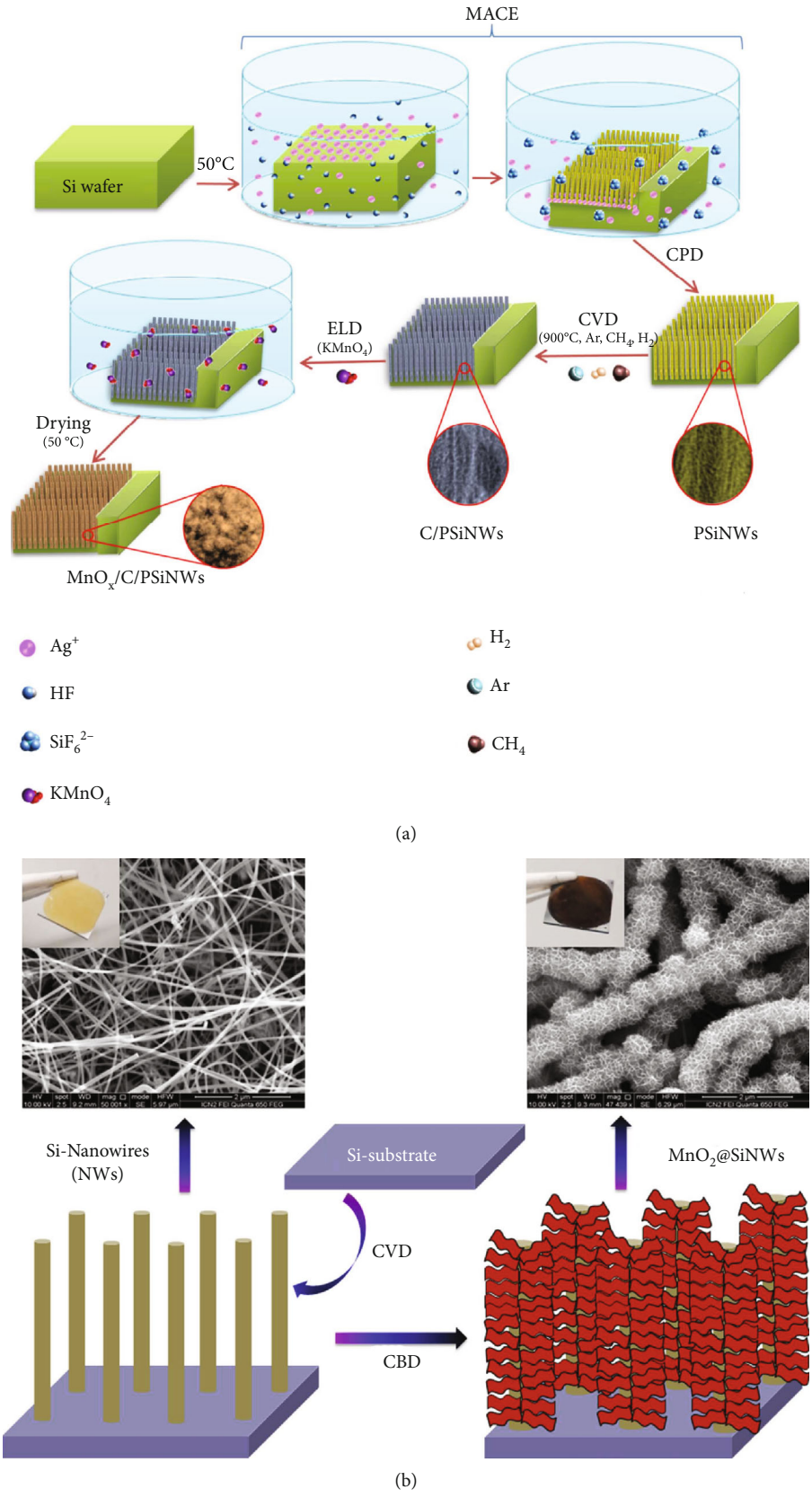


FIGURE 2: (a) Schematic illustration of the composite fabrication based on carbonized porous silicon nanowires and MnO_x decorated carbonized porous silicon nanowires. Reproduced with permission from Ref. [41]. Copyright 2017, Royal Society of Chemistry. (b) Schematically introduction of the fabrication process for hierarchical MnO_2@silicon nanowires. Reproduced with permission from Ref. [42]. Copyright 2015, Springer Nature.

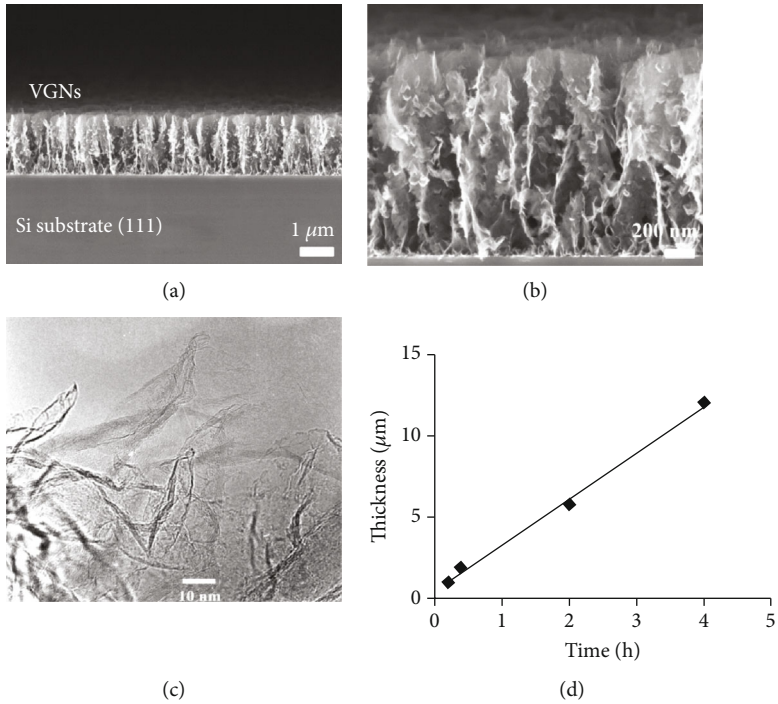


Figure 3: (a, b) SEM images and (c) TEM image of vertically oriented graphene nanosheets deposited onto doped Si substrates, (d) the thickness of the graphene under different time. Reproduced with permission from Ref. [47]. Copyright 2015, Royal Society of Chemistry.

double-layer capacitors [51]. After the deposition of nano-nickel, the silicon-based microchannel plate can be stable in most electrolytes. Compared to bare Si-based microchannel plate, the three-dimensional nano-Ni electrode based on Si-based microchannel plate has achieved increased specific capacitance due to the enhanced electron/ion transport derived from porous structure and three-dimensional architecture. Layered Si-based nanosheets have been obtained and used as electrode materials in supercapacitors with the voltage of 4 V [52]. The Si-based nanosheets exhibited excellent electrochemical properties including high areal specific capacitance (4.43 mF cm^{-2}), outstanding volumetric energy density (7.65 mWh cm^{-3}), and power density (9312 mW cm^{-3}), as well as stable cycling stability over 12000 cycles.

2.3. Si Nanoparticles. Si nanoparticles have been potential candidates for high-performance supercapacitors. Liu et al. have dispersed ultrasmall silicon nanoparticles in polyaniline to form a composite Si/PANI material as the active material for supercapacitors [53]. For the composite Si/PANI, silicon nanoparticles and polyaniline provided electrode double-layer capacitance and pseudocapacitance, respectively. Electrochemical characterizations of the composite Si/PANI displayed superior performances in terms of high power density of 220 kW kg^{-1}, energy density of 30 Wh kg^{-1}, and good cycling stability over 1000 charging/discharging times.

Gardner et al. have demonstrated electrochemical capacitors consisting of porous silicon nanostructures with very high surface-to-volume ratios [54]. Nanopore morphologies and passivation coatings played important role in maximizing energy and power densities. Besides, enhanced electrochemical properties can be achieved through atomic layer deposition (ALD) titanium nitride or chemical vapor deposition (CVD) carbon coatings.

A unique conductive bacterial cellulose composite with silicon nanoparticles has been described using bacterial cellulose as a template for binding silicon nanoparticles [55]. Followed the in situ polymerization process, a conformal coating of polyaniline was formed on the modified silicon nanoparticles to prepare composites. The composites featured with flexible and stable electrical conductivity for electrochemical applications. Ternary hybrid materials Fe_3O_4/carbon-coated Si have been fabricated with enhanced electrochemical performances [56]. The carbon layers were grown on the surface of Si by the thermal vapor deposition, which not only afford anchoring sites for the deposition of Fe_3O_4 but also improve the contact between Si and reactive electrolyte. The deposition of Fe_3O_4 nanoparticles offers pseudocapacitive behavior for the hybrid materials to achieve enhanced electrochemical performances.

2.4. Si 3D Structure. Three-dimensional structures are favorable to boost the capacitive behaviors of Si-based supercapacitors due to their large surface-to-volume ratio and more available active sites for enhanced charge transfer. For example, three-dimensional silicon microchannel plates have been used to construct an electronic double-layer supercapacitor, wherein the 3D structure served as substrates for the deposition of positive active MnO_2 nanoflakes [57]. Benefited from the 3D structure, increased active sites render superior electrochemical performances in the organic electrolyte to that of a planar structure.

Figure 4 displays the morphologies of the samples Ni/Si-MCPs and MnO_2/Ni/Si-MCPs, wherein Ni/Si-MCPs

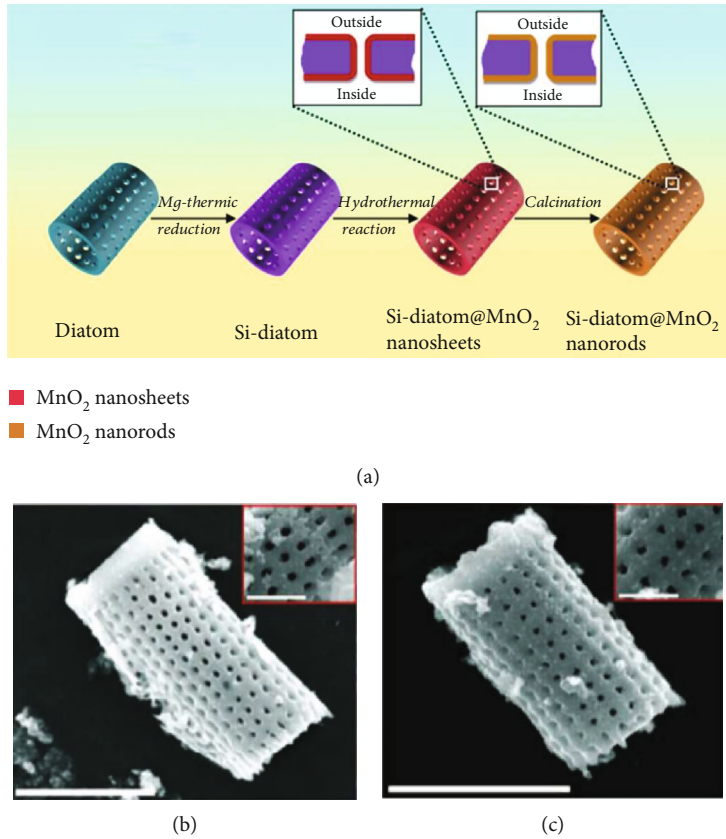


FIGURE 4: (a) The schematic display of the fabrication of silicon-diatom@MnO_2 electrodes. (b) SEM image of silica before conversion and (c) SEM image of silica diatom after conversion. Reproduced with permission from Ref. [60]. Copyright 2017, Royal Society of Chemistry.

meaned three-dimensional nickel/silicon microchannel plates and MnO_2/Ni/Si-MCPs were prepared after electrodeposition of MnO_2 on Ni/Si-MCPs [58]. Nickel covers on Ni/Si-MCPs effectively decrease the internal resistance up to less than 1 Ω. Three-dimensional structures allowed adequate sites for facilitated ion diffusion. The presence of intertwined MnO_2 nanoflakes was deposited on Ni/Si-MCPs densely and largely contributed to the capacitance. Microsupercapacitors based on MnO_2/Ni/Si-MCPs have been measured with the capacitance of 0.961 F cm^{-2} (323.1 F g^{-1}) and 91.1% retention after 1000 cycles, demonstrating the robustness and durability in a neutral 1 M Na_2SO_4 solution.

Hybrid $Ba_{0.65}Sr_{0.35}TiO_3$(BST)/$NiSi_2$/silicon microchannel plate has been fabricated as supercapacitor electrodes by combining with the sol-gel method and silicon microfabrication technology [59]. An electrochemical etching silicon microchannel plate (MCP) was utilized as a backbone of the 3D architecture, and followed nickel layer was deposited on the surface. A layer of $Ba_{0.65}Sr_{0.35}TiO_3$ was synthesized by the sol-gel technique in conjunction with multiple steps of vacuum spin-coating onto the nickel-coated silicon MCP which serves as current collector. The supercapacitor with 3D structure exhibits double-layer capacitance characteristic with a maximum capacitance (784 F g^{-1}). The capacitance displayed a slight decrease with loss of 6.3% after 700 charging/discharging cycles. It resulted from the presence of the 3D structure which favors for faster ion conductivity compared to the 2D planar structure.

Unique 3D silicon-diatom@MnO_2 electrodes have been fabricated, wherein 3D silicon-diatoms were conversed from diatomites (SiO_2) via the magnesiothermic reduction method and MnO_2 nanosheets were deposited onto 3D silicon-diatoms [60], as illustrated in Figure 4(a). Figures 4(b) and 4(c) display the SEM images of silica before and after the magnesiothermic reduction method. Benefited from the synergic effects between silicon-diatoms and MnO_2 layers, excellent electrochemical properties were investigated with a high specific capacitance of 341.5 F g^{-1} at 0.5 A g^{-1}, good rate capability (47.7% retention at the current density of 10 A g^{-1}), and outstanding cycling stability over 2000 cycles).

Ruthenium oxide (RuO_2) has been combined with Si current collector to fabricate the three-dimensional RuO_2/Si configuration [61]. Si current collectors served as templates for the deposition of tubular RuO_2. Due to the advantages of Si, Si/RuO_2 were assembled on a chip symmetric supercapacitor with a high areal capacity of 23 mF cm^{-2} at 10 mA cm^{-2}. The tubular morphology and pseudocapacitive behavior of RuO_2 rendered the single electrode to perform a specific capacitance of 99.3 mF cm^{-2} and 70 F g^{-1} at 5 mV s^{-1} and satisfied cyclability in neutral Na_2SO_4 solution.

Innovative 3D symmetric microsupercapacitor has been fabricated using polypyrrole- (PPy-) coated silicon nanotree (SiNTr) as hybrid electrodes [62]. The PPy coating was deposited via an electrochemical method onto the surface of SiNTr electrodes which were grown on silicon substrates by chemical vapor deposition. The hybrid microsupercapacitor device

illustrated excellent electrochemical performance benefited from the synergy between the pseudocapacitive behavior of PPy and electric double-layer capacitive behavior of SiNWs in the aprotic ionic liquid (N-methyl-N-propylpyrrolidinium bis(trifluoromethylsulfonyl)imide; $PYR_{13}TFSI$). Microsupercapacitors based on PPy/SiNTr revealed excellent electrochemical properties including a high specific capacitance ($14\,mF\,cm^{-2}$), a high energy density ($15\,mJ\,cm^{-2}$) at the voltage of 1.5 V, and outstanding cycling stability with a loss of approximately 30% after thousands of galvanostatic charge-discharge cycles.

Porous silicon has been considered as candidates towards microsupercapacitor devices due to its convenient integration. However, the power efficiency of pristine porous silicon was limited by the poor wettability and poor chemical stability in electrolytes and the high resistance. To modify the surface of nanoporous silicon, graphene with interconnected networks has been adhered to nanoporous silicon matrix by electrochemical polymerization of 2,6-dihydroxynaphthalene and following thermal treatment [63]. The graphene/silicon can be easily assembled for solid-state supercapacitor devices via scalable technology. The symmetric supercapacitors displayed remarkable properties especially enhanced capacity retention at ultrahigh rates. An ultrathin layer of titanium nitride has been coated by atomic layer deposition onto the porous silicon matrix (PS-TiN) to achieve excellent wettability of Si-based electrodes in aqueous and organic electrolytes [64]. The PS-TiN nanostructures were fabricated in a chip monolithically and provided outstanding performances such as a high specific capacitance of $15\,F\,cm^{-3}$, a high energy density of $1.3\,mWh\,cm^{-3}$, and power density up to $214\,W\,cm^{-3}$ as well as remarkable stability over 13000 cycles.

2.5. Si-Doped Materials. Silicon-doped carbon-based materials have been demonstrated with excellent properties and extended application. Silicon-doped carbon-based materials are promising to be used as an active material for supercapacitors. Silicon doping can modulate the electronic structure and improve its physical/chemical properties. There are some reports about Si-doped carbon for application in supercapacitors.

Ramasahayam et al. have demonstrated the synthesis of N/P/Si tri-doped C (NPSiDC) through a simple, rapid, and economical one-pot microwave-assisted method [65]. The as-prepared electrode materials exhibited tremendous potential for supercapacitor applications with the highest specific capacitance value of $318\,F\,g^{-1}$ and electrochemically stability after 2000 cycles in 6 M KOH.

Si-doped reduced graphene oxide (Si-rGO) has been synthesized through annealing treatment of triphenylsilane and graphene oxide [66]. Figure 5 characterizes the morphology and doping components of Si-rGO. The TEM and SEM images illustrated two-dimensional geometric transparent graphene sheets with crumpled structures after the reduction process. Energy dispersive X-ray (EDX) spectrum was applied to confirm the doping components and distribution of Si. The homogeneous Si-doping on graphene was favorable for superior electrochemical ability to the pristine graphene. Supercapacitors based on Si-rGO exhibited significant enhancement in electrochemical properties. The specific capacity is increased by 48.5%. This suggests that silicon doping can effectively improve the electrocatalytic ability and electrochemical performance.

Song et al. have illustrated the preparation of multidimensional nanocarbons hybridized with mesoporous SiO_2 via a facile and versatile strategy [67]. Mesoporous SiO_2 on nanocarbons were revealed as potential candidates for supercapacitors. With the coating of SiO_2, the nanocarbons achieved increased surface area and low internal resistance. Excellent electrochemical performances were revealed with a high specific capacitance ($23.84\,mF\,cm^{-2}$ at $20\,mV\,s^{-1}$).

Tri-heteroatom- (N, P, Si-) doped nonporous carbon nanofibers were synthesized by using tetraethyl orthosilicate, phosphorus acid, and polyacrylonitrile as the source of Si, P, and N, respectively [68]. The heteroatom-doped carbon is a feasible and ecofriendly electrode material for high-performance supercapacitors with high gravimetric capacitance ($243.7\,F\,g^{-1}$ at $0.5\,A\,g^{-1}$), rate capability (83% retention at $30\,A\,g^{-1}$), and stable charging/discharging cycling life. Particularly, it possessed a high packing density with a maximum volumetric capacitance of $253.4\,F\,cm^{-3}$ at $0.5\,A\,g^{-1}$.

P-doped silicon (P-Si) with graphene coating has been demonstrated as a promising electrode in grid-scale and integrated electrochemical energy storage [69] with a wider electrochemical window. Wu et al. have demonstrated passivated porous silicon few-layer graphene sheets in an electrochemical double-layer supercapacitor [70]. The hybrid structure was investigated with optimized capacitive performances with a high areal capacitance ($6.21\,mF\,cm^{-2}$ at $1000\,mV\,s^{-1}$) and an unusual cyclic stability of 131% after 10000 cycles.

2.6. Large-Voltage Microsupercapacitors. Energy and power density are directly proportional to the capacitance and the square of the operating voltage as follows (C represents the capacitance, V represents the operating voltage, and ESR represents the equivalent series resistance).

$$E = 0.5CV^2,$$
$$P_{max} = \frac{V^2}{4ESR}. \quad (1)$$

Besides the charge storage abilities of active materials, the maximum operation voltage is also directly determined by the energy density and power density. A large-voltage window electrolyte is effective to obtain enhanced electrochemical performances with the open voltage range due to the quadratic relationship.

EMI-TFSI electrolytes have been illustrated as ionic media for the cell voltage up to 4 V [71]. The supercapacitor has been fabricated based on two-dimensional Si-based nanosheets resulting from oxidation and exfoliation of $CaSi_2$. These silicon-based nanosheets with two-dimensional structure generated high surface areas for favorable ion diffusion. The electrochemical properties have been investigated in Figure 6 by using the cyclic voltammetric (CV) and

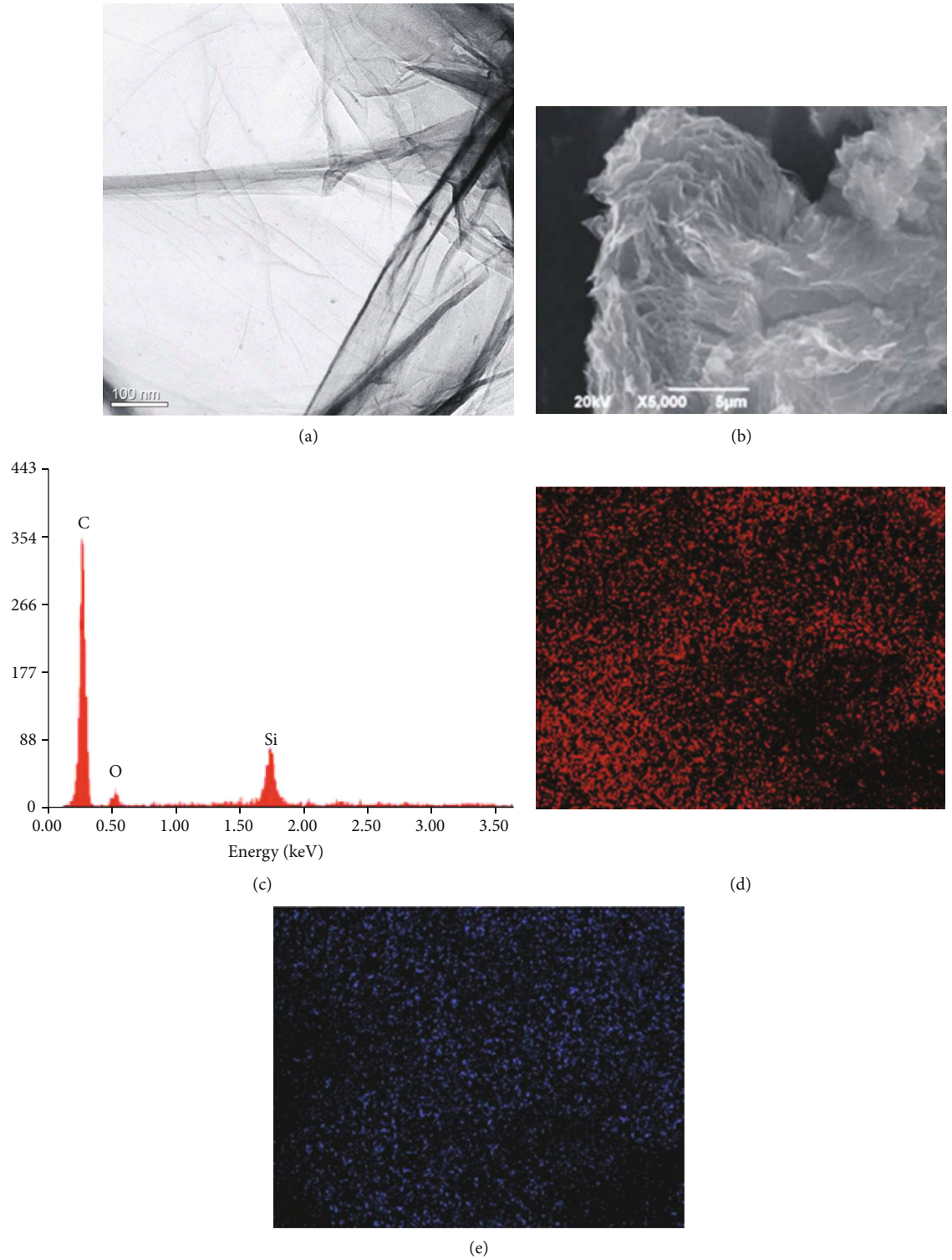


Figure 5: The morphology and doping components of Si-doped reduced graphene oxide. (a) TEM image, (b) SEM image, (c) EDX spectrum, (d) the mapping distribution of C element, and (e) the mapping distribution of Si element. Reproduced with permission from Ref. [66]. Copyright 2016, Royal Society of Chemistry.

galvanostatic charge-discharge (GCD) measurement. The CV testing was conducted as shown in Figure 6(a) with a nearly rectangular shape under various cell voltage, indicating the EDLC behaviors.

The CV measurements as shown in Figures 6(b)–6(d) were conducted at different scan rates and performed nearly symmetrical rectangular which matched the charge storage mechanism of the electrical double-layer capacitor. Similar

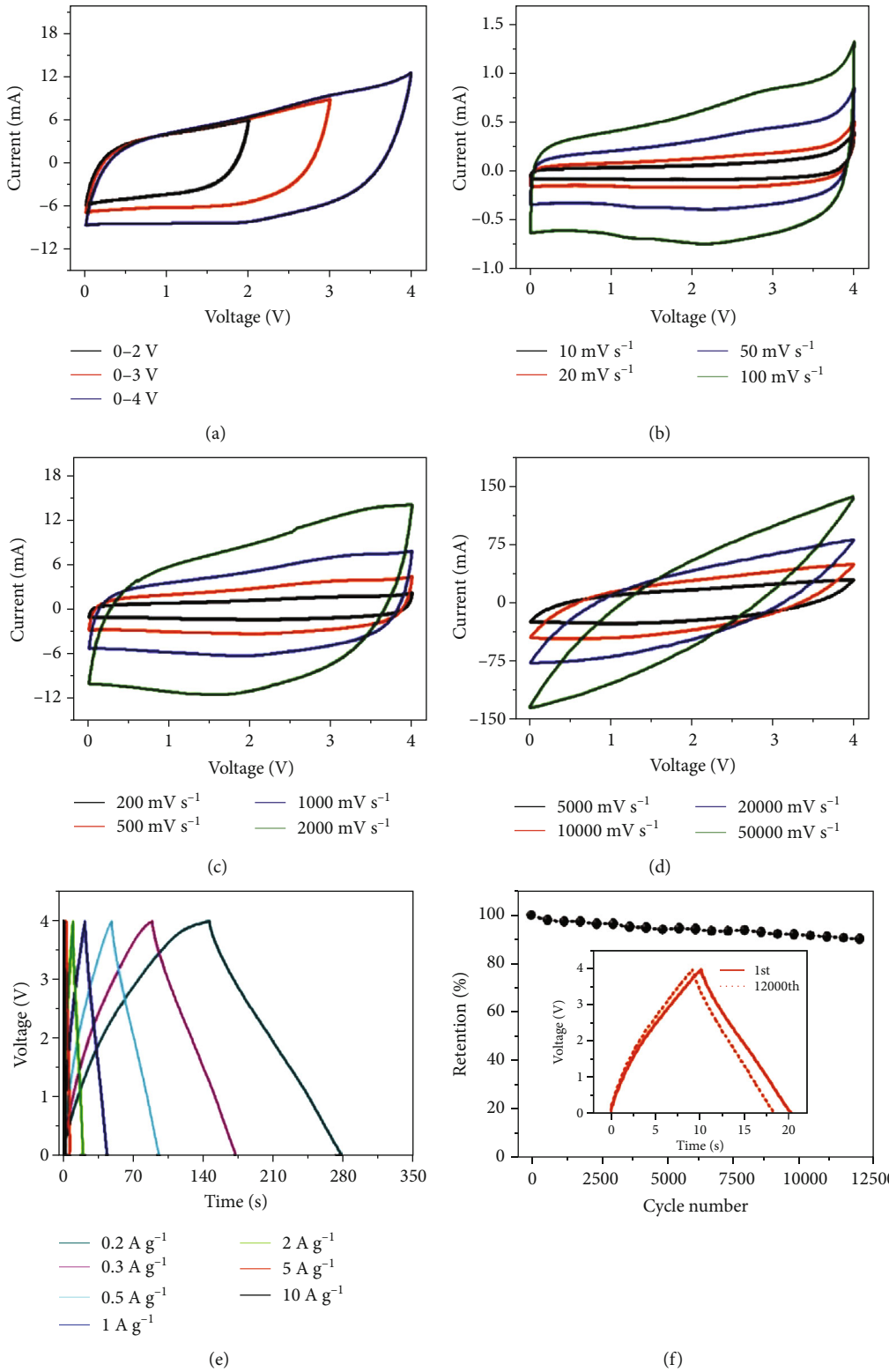


Figure 6: Electrochemical performances of two-dimensional Si-based nanosheet electrode in the ionic electrolyte using EMI-TFSI as the media. (a) CV curves at the scan rate of 1000 mV s^{-1} with the cell voltage ranging 2.0 to 4.0 V. (b) CV curves at different scan rates from 10 to 100 mV s^{-1}. (c) CV curves at different scan rates from 200 to 2000 mV s^{-1}. (d) CV curves at various scan rates from 5000 to 50000 mV s^{-1}. (e) GCD curves at various current densities from 0.2 to 10 A g^{-1}. (f) Cycling stability over 12000 charging/discharging process at 2 A g^{-1}. Reproduced with permission from Ref. [71]. Copyright 2020, Wiley-VCH.

CV curves were investigated even though with high scan rates of 5000-50000 mV s^{-1}. The corresponding specific capacitances were calculated with 4.43 mF cm^{-2} at a scan rate of 10 mV s^{-1} and 2.49 mF cm^{-2} at a scan rate of 2000 mV s^{-1}. The GCD curves in Figure 6(e) were tested under different current densities to verify the results of 1CV measurement. In addition, the cycling performance of the electrode was examined at 2 A g^{-1} as displayed in Figure 6(f). The inset displayed outstanding electrochemical reversibility and cycling stability through the comparison of the capacitive behaviors after 12000th charge/discharge cycling.

The work voltages are different for various types of electrolytes and play a vital role in the energy density and power density of supercapacitors. Aqueous electrolytes and organic electrolytes can exhibit the stable electrochemical behaviors at 1.3 V and 2.7 V, respectively. Instead, ionic liquid electrolytes have been widely explored as alternative electrolytes for their high thermal stability (>300°C) and broad open voltage range (>4 V) in Si-based supercapacitors. Aradilla et al. constructed symmetric microsupercapacitor in an protic ionic liquid (N-methyl-N-propylpyrrolidinium bis(trifluoromethylsulfonyl)imide) (PYR_{13}TFSI) electrolyte [72]. The devices operated at a wide voltage window of 4 V, and the Si-based supercapacitors delivered the power density of 182 mW cm^{-2} and the energy density of 190 μJ cm^{-2}. Berton et al. have investigated an ionic liquid electrolyte (EMI-TFSI) for large-voltage window SiNW-based microsupercapacitors [73]. An extended operating voltage range (up to 4 V) can be obtained and enables with a promising power density of 472 μW cm^{-2}. It was noted that the presence of ionic liquid electrolyte soluted the etching of Si-based electrode materials in common aqueous or organic electrolytes under a wide voltage.

Another promising approach to widen the cell voltage of supercapacitors is to develop asymmetric supercapacitors, which consist of a positive electrode based on pseudocapacitive or battery mechanism and a negative electrode based on electrode double-layer behavior. Outstanding charge storage capacity can be investigated due to the synergistic contributions between the electric double-layer capacitive contribution and the pseudocapacitive contribution. For example, an asymmetric supercapacitor has been fabricated via compact integration of fullerene-like carbon decorated carbon nanotubes (FC-CNTs) regulated by the employed dry-etched silicon taper nanorod (Si-TNR) scaffold [74]. The on-chip microsupercapacitors represent the capability of rapid energy storage (192 mF cm^{-2} at a scan rate of 1 mV s^{-1}, 123 mF cm^{-2} at a scan rate of 1000 mV s^{-1}).

In particular, Li-ion capacitors (LIC) have been proposed with a higher voltage and a considerably higher energy density than other existing capacitor systems [75]. Liu et al. have manufactured a high voltage of LIC between Si/AC and AC/$LiMn_2O_4$ [76]. The electrode materials are synthesized by a microdome-patterned process to control the shape of the current collector and electrode material. The Li-ion capacitors have been investigated as candidates to broaden the working voltage window for enhanced energy density and power density.

The electrochemical performance of the supercapacitor is also directly determined by ionic media in the electrolytes. Kim et al. have fabricated asymmetric supercapacitors in various ionic media with excellent capacitive performances and energy density, for example, a solid-state asymmetric supercapacitor based on $MgCo_2O_4$ nanoneedles onto porous silicon carbide flakes (SiCF). The solid-state asymmetric supercapacitor was constructed by SiCF/$MgCo_2O_4$//SiCF in the electrolyte system of PVA-KOH-KI [77, 78]. The electrode demonstrated an outstanding specific capacitance (185.88 C g^{-1}) and energy density of (41.308 Wh kg^{-1}) and cycling stability.

In addition, PVA-KOH-p-nitroaniline (PVA-KOH-PNA) gel electrolyte has been employed as a redox active electrolyte in a quasi-solid-state flexible asymmetric supercapacitor. For example, Kim et al. have fabricated a quasi-solid-state flexible asymmetric supercapacitor in PVA-KOH-PNA gel electrolyte using micro- and mesoporous SiC/Fe_3O_4 composites as the electrode materials [79]. The SiC/Fe_3O_4 flexible solid-state asymmetric supercapacitor displayed superior electrochemical properties including a high specific capacitance (97.6 F g^{-1} at 5 mV s^{-1}) and a maximum energy density (48.94 Wh kg^{-1} at the power density of 463.64 W kg^{-1}). Similarly, their group has assembled a novel solid-state asymmetric supercapacitor device wherein the SiCF/$MgCo_2O_4$ and the SiCF/Fe_3O_4 served as the positive electrode and the negative electrode in the PVA-KOH-PNA gel electrolyte [80]. The hybrid nanostructure electrodes exhibited outstanding electrochemical properties such as a high specific capacitance (161.77 F g^{-1} at 5 mV s^{-1}) and a maximum energy density (72.79 Wh kg^{-1} at the power density of 727.96 W kg^{-1}).

An optimal electrolyte has been prepared and used as electrolytes for symmetric microsupercapacitors with a large and stable cell voltage of 3.5 V and wide operating temperatures [81]. The electrolyte is derived from the mixture of propylene carbonate (PC) and N_{1114}TFSI ionic liquid (50 : 50% w.t). The device in the optimal electrolyte performed comparable properties to those of pure ionic liquid. The device illustrated a high areal capacitance (150 mF cm^{-2}), a high energy and power density (1 mJ cm^{-2} and 16 mW cm^{-2}), and remarkable cycling stability over $3 * 10^6$ galvanostatic charging/-discharging cycles under room temperature. Hybrid devices based on thin atomic layer deposition coatings have been constructed to obtain the larger cell voltage [82]. A thin alumina layer on Si-based materials can prevent the oxidation of Si-based electrodes for enhanced reproducible and stable electrochemical behaviors. The as-fabricated symmetric devices performed a large cell voltage of 6 V in the EMI-TFSI electrolyte with an outstanding cyclability over 10^6 charge/discharge cycles.

3. SiC Electrode Materials

Silicon carbide (SiC) materials have attracted a great deal of attention in supercapacitors especially on-chip microsupercapacitors as electrode materials due to the high conductivity, high surface area, effective electrolyte transport and high thermal and chemical stability, long life time, and high-temperature operations [83]. Moreover, silicon

carbide materials are compatible with standard microfabrication techniques, which render them beneficial for fabrication and integration as on-chip Si-based microsupercapacitors.

Alper et al. have reported silicon carbide nanowires as electrode materials in an aqueous electrolyte for microsupercapacitors [84]. The SiC nanomaterials here are less than 10 mm thick and can withstand $2 * 10^5$ charge/discharge cycles in an aqueous electrolyte while maintaining over 95% of their initial capacitance value.

Silicon carbide flakes (SiCFs) with micro/mesopores were obtained by following carbonization of waste Si wafer [85, 86]. During the carbonization process, the micropores resulted from the partial evaporation of Si atoms in waste Si wafer and the mesopores occurred from the integration of neighboring micropores. Silicon carbide flakes featured with a high surface area ($1376\,m^2\,g^{-1}$) without any chemical or physical activation and constructed as electrodes in two-electrode supercapacitor cells with aqueous and organic electrolytes. High-charge storage capacities have displayed the specific capacitance value of $49.2\,F\,g^{-1}$ and $38.7\,F\,g^{-1}$ at $5\,mV\,s^{-1}$ in an aqueous and organic electrolyte, respectively.

Moyano et al. have developed a silicon carbonitride-graphene oxide hybrid material through a preceramic polymer route [87]. The structures are tested as electrodes for supercapacitors, reaching a gravimetric capacitance of $39\,F\,g^{-1}$ that remains stable after 7000 charge/discharge cycles.

Silicon carbide spheres with hierarchical pore structures have been employed to construct symmetrical supercapacitor in the aqueous electrolyte (1 M potassium chloride, KCl) and the organic electrolyte (1 M tetraethylammonium tetrafluoroborate in acetonitrile, $TEABF_4$/AN) [88]. Hierarchical pore structures consisted of micropores and mesopores interconnected. Micropores offered higher specific capacitance $60.3\,F\,g^{-1}$ at $5\,mV\,s^{-1}$ in the aqueous electrolyte more than that in the organic electrolyte. Mesopores offered the higher specific capacitance in the organic electrolyte benefited from the mesopores allowed for the effective ion diffusion of organic media. Besides, the wide operating voltage of the organic electrolyte rendered the energy density a high value of $102.59\,W\,h\,kg^{-1}$ at $5\,mV\,s^{-1}$, which is more than three times of the energy density in aqueous electrolytes.

Silicon carbide nanowire-based composite materials have been prepared by the growth of SiC nanowire on carbon fabric [89]. The as-prepared SiC nanowire/carbon fabric materials can be directly used as the EDLC electrode with no binder, wherein flexible carbon fabric served as a conductive and stable substrate. Electrochemical performances were revealed with a high areal capacitance ($23\,mF\,cm^{-2}$ at $50\,mV\,s^{-1}$) and noticeable cycling stability after 10^5 cycles at room temperature. Besides, silicon carbide nanowires on carbon fabric performed excellent electrochemical properties for high temperature. The capacitances increased with the rise of the working temperature, and 90% of the initial capacitance was retained after 10^5 cycles even the measurement temperature of 60°C.

Mesoporous silicon carbide/carbon (SiC/C) materials have been synthesized via a self-templating method [90]. The synthesis utilizes salt byproducts as the internal self-forming templates. When using as the supercapacitor electrode, the SiC/C composite exhibited outstanding electrochemical properties such as a specific capacitance ($220\,F\,g^{-1}$), stable long cycling stability, and a good rate capability.

Metal oxides/hydroxides have been proved to be promising electrode materials due to their redox pseudocapacitive mechanism for fast redox kinetics and higher specific capacitance. Among these metal oxide materials, MnO_x and Fe_3O_4 have attracted considerable focus and widely used because of their intrinsic advantages such as environmental friendliness, natural abundance, and low cost [91]. Composites consisting of SiC and MnO_x or Fe_3O_4 have been reported in recent years.

For example, Kim et al. suggested the use of microsphere silicon carbide/nanoneedle MnO_2 (SiC/MnO_2) composites in supercapacitors [92]. SiC was treated with hydrogen peroxide to obtain oxygen-containing functional groups as anchoring sites on the surface for the deposition of MnO_2. The introduction of MnO_2 resulted in the enhanced capacitive properties with the specific capacitance of $273.2\,F\,g^{-1}$ at a scan rate of $10\,mV\,s^{-1}$ in the three-electrode devices in the aqueous electrolyte (1 M Na_2SO_4). Birnessite-type MnO_x was also composited to silicon carbide microsphere and the composites denoted as SiC/B-MnO_x [93]. The SiC/B-MnOx electrodes were measured with an optimized specific capacitance ($251.3\,F\,g^{-1}$ at $10\,mV\,s^{-1}$) in 1 M Na_2SO_4 electrolyte. Silicon carbide/MnO_2 nanoneedle composites have been developed as the positive electrode materials in the asymmetric electrochemical supercapacitor in neutral aqueous Na_2SO_4 solution [94]. When activated carbon is applied as the negative electrode materials, the asymmetric devices performed the operating voltage up to 1.9 V.

Composites consisting of SiC and Fe_3O_4 have been synthesized through the chemical deposition of Fe_3O_4 on micro- and mesoporous SiC flakes [95]. The SiC/Fe_3O_4 electrode displayed a high specific capacitance ($423.2\,F\,g^{-1}$ at a scan rate of $5\,mV\,s^{-1}$) and a notable rate performance (81.8% from 5 to $500\,mV\,s^{-1}$) in the aqueous electrolyte using 1 M KOH as media. The outstanding performances benefited from the electric double-layer capacitive contribution of the SiC and the pseudocapacitive contribution of the Fe_3O_4 nanoparticles.

$Ni(OH)_2$ has attracted much attention due to its excellent electrochemical activity. SiC nanowires@$Ni(OH)_2$@carbon fabric have been composited via the chemical vapor deposition (CVD) and following electrochemical cathodic deposition method [96]. SiC nanowires@$Ni(OH)_2$ were grown on flexible carbon fabric and directly used as the electrode to fabricate a solid-state supercapacitor. The SiC nanowires@-$Ni(OH)_2$@carbon fabric electrode revealed good flexibility, high areal capacitances of $1724\,F\,g^{-1}$, and stable cyclability properties. The above-mentioned SiC-based materials have been illustrated in Table 1 in detail.

Silicon carbide nanofiber membranes have been synthesized by solution blowing and calcination process and pressed onto Ni foam directly as electrodes in

Table 1: SiC-based materials for supercapacitors.

Materials	Synthesis	Morphology	Performance	Ref.
SiC	Low-pressure chemical vapor deposition	Nanowire	240 mF cm^{-2} at 100 mV s^{-1}	[84]
SiC	One-step carbonization	Nanoflakes	49.2 F g^{-1} at 5 mV s^{-1}	[85, 86]
SiC	Self-assembly	Spheres	82.9 F g^{-1} at 5 mV s^{-1}	[88]
SiC/C	Chemical vapor deposition	Nanowire	23 mF cm^{-2} at 50 mV s^{-1}	[89]
SiC/C	Self-templating method	Fiber	220 F g^{-1}	[90]
SiC/MnO_2	Deposition	Microsphere/nanoneedle composites	273.2 F g^{-1} at 10 mV s^{-1}	[92]
SiC/MnO_2	Deposition	Composites	251.3 F g^{-1} at 10 mV s^{-1}	[93]
SiC/Fe_3O_4	Chemical deposition	Composites	423.2 F g^{-1} at 5 mV s^{-1}	[95]
SiC/$Ni(OH)_2$	Chemical vapor deposition	Nanowires	1724 F g^{-1} at 2 A g^{-1}	[96]

supercapacitors [97]. SiC-NFMs with high productivity exhibited good electrochemical performances in supercapacitors such as the enhanced specific capacitance of 189 F g^{-1} and remarkable cyclability with 91.7% retention after 3000 cycles.

SiC have been deposited onto porous alumina substrates (AAO) with Ag coating via DC magnetron cosputtering technique at room temperature and used as binder-free electrodes to fabricate symmetric supercapacitors [98]. The SiC@Ag-AAO symmetric supercapacitor device operated the cell voltage of 1.8 V and delivered high energy density of 31.43 Wh kg^{-1}) and maximum power density of 18.8 kW kg^{-1} at 17.76 Wh kg^{-1} with stable cycling ability over 3000 cycles.

Three-dimensional silicon carbide-based frameworks have been prepared by employing a template method and subsequent carbonization via an aerosol spray drying method [99–101]. Hierarchical micro-, meso-, and macroporous structures were interconnected and contributed to the enhanced electrochemical performances. Briefly, micropores can increase charge accommodation. Mesopores offered more accessible active sites for ion diffusion. Macropores allowed shorter charging/discharging time. Consequently, enhanced electrochemical performances were investigated for three-dimensional silicon carbide-based frameworks in the aqueous electrolyte (1 M Na_2SO_4) and the ionic-liquid electrolyte (3-ethyl-3-methylimidazolium bis(trifluorosulfonyl)imide).

It is reported that the oxygen-containing functional groups in porous SiC exerted nonnegligible influence on the capacitive behavior [102]. Kim et al. have investigated the electrochemical properties of oxidized micro- and meso-SiC flakes wherein the oxygen-containing functional group was introduced during the charge/discharge process. The oxidized SiC-based electrodes performed superior specific capacitance (243.3 F g^{-1} at 5 mV s^{-1}) to that of SiC-based electrodes due to the pseudocapacitive contribution resulted from the presence of oxygen functional group.

4. Conclusions

In this review, we have summarized the development of supercapacitors consisting of Si-based materials and their derivatives. To illustrate the active electrode materials, aspects of the representative progresses of Si-based materials and their derivatives have been summarized on synthesis, properties, surface modification, and electrochemical properties. The capacitive performances are related to the morphology and structure of electrode materials. Hence, a variety of electrode materials with different morphology and structures have been outlined as follows: (1) Si nanowire, (2) Si nanosubstrates, (3) Si nanoparticles, (4) Si 3D structure, and (5) Si-doped materials. Meanwhile, the overall behaviors in microsupercapacitor application have been illustrated in terms of specific capacitance, rate capability, cycling life, and energy density. Furthermore, large-voltage microsupercapacitors are candidates for next-generation wearable, portable, and stretchable devices. The electrolyte optimized for Si-based supercapacitors to obtain enhanced energy density. These encouraging achievements demonstrate the potential applications of Si-based materials and their derivatives for high-performance supercapacitors.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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