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Review

CNTs-Supercapacitors: A Review of Electrode Nanocomposites Based on CNTs, Graphene, Metals, and Polymers

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Abstract: Carbon nanotubes (CNTs), due to mechanical, electrical, and surface area properties and their ability to adapt to different nanocomposite structures, are very substantial in supercapacitor electrodes. In this review, we have summarized high-performance, flexible, and symmetry CNT supercapacitors based on the CNTs/graphene, CNTs/metal, and CNTs/polymer electrodes. To present recent developments in CNT supercapacitors, we discuss the performance of supercapacitors based on electrical properties such as specific capacitance (SC), power and energy densities, and capacitance retention (CR). The comparison of supercapacitor nanocomposite electrodes and their results are reported for future researchers.

Keywords: supercapacitor; CNTs; metal; graphene; PANI; specific capacitance

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1. Introduction

Supercapacitors are capable energy storage systems, as they offer fast charge/discharge, high cycle stability, high power density, and stable electrical properties. Supercapacitors in several industries are used for energy harvesting and high-power systems in electric vehicles. Recently, supercapacitors based on CNTs composite electrodes have received more attention due to the electrical and chemical properties of CNTs [1–7]. Growth in the publication of CNT-based supercapacitors is shown in Figure 1.

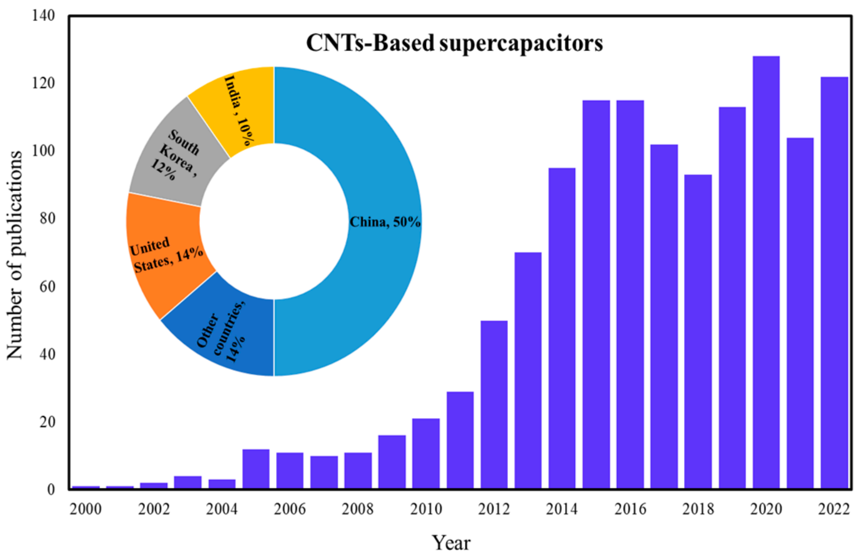

Figure 1. Growth in CNT-based supercapacitor publications.

The SC is a crucial parameter in the calcification of supercapacitors. The SC can be obtained by cyclic voltammetry and charge/discharge methods. From the cyclic voltammetry method, the SC can be obtained from the following formula:

$$C_m = \frac{\int_{V_1}^{V_2} I\, dV}{m\, S\, \Delta V} \tag{1}$$

where *m* is the mass of the active materials' electrode, S is the voltage scan rate (mV/s), ΔV is the potential window, *I* is the current in the CV curve, and *m* is the mass of the electrode materials. From the charge/discharge method, the SC can be obtained from the following formula [8]:

$$C_m = \frac{I}{\left(m\,\frac{dV}{dt}\right)} \tag{2}$$

Figure 2 shows the SC of CNT-based supercapacitors that was published in 2022. As shown in Figure 2, 14% of supercapacitors achieved a SC higher than 1000 F/g and 65% lower than 500 F/g.

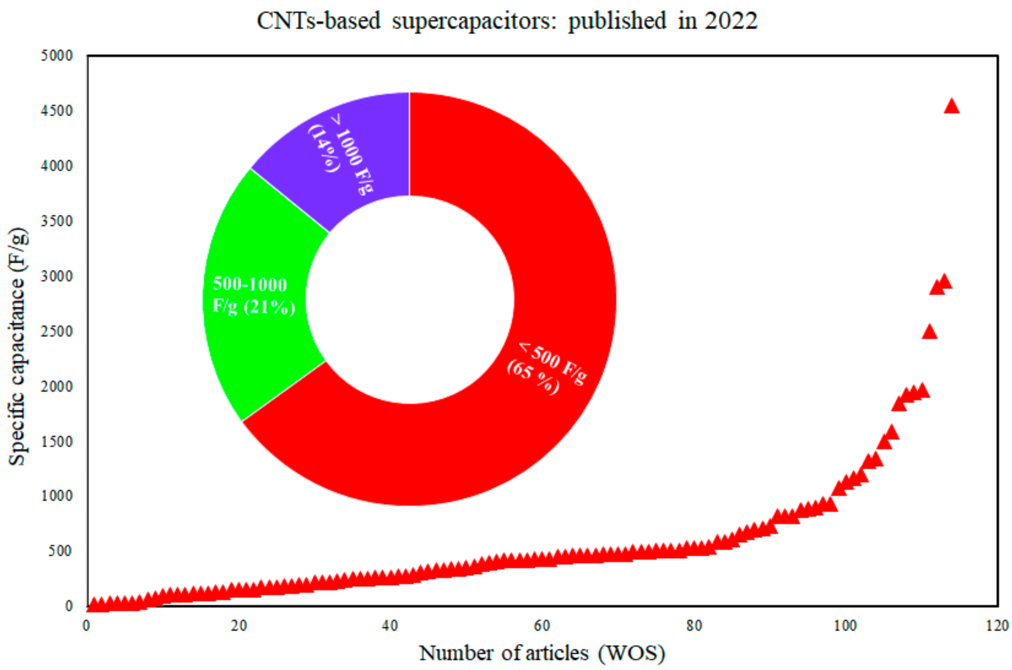

Figure 2. Specific capacitance of CNTs–based supercapacitors as a function of the number of articles.

The co-occurrence keywords analysis of the CNT-based supercapacitor by VOSviewer from the Scopus for 2022 publications is shown in Figure 3. As can be observed from Figure 3, the electrode nanocomposite based on the CNTs/graphene, CNTs/metal, and CNTs/polymer are more highlighted in the co-occurrence keywords analysis. Other characterizations such as symmetric [9–11], flexible [12,13], wearable electronic [14,15], cellulose [16–18], cycling stability [19,20], pseudocapacitor [21,22], solid-state supercapacitor [23–25], activated carbon [26–28], polymers [29–31], MXene [32,33], and CNTs [34–46] were mentioned in Figure 3.

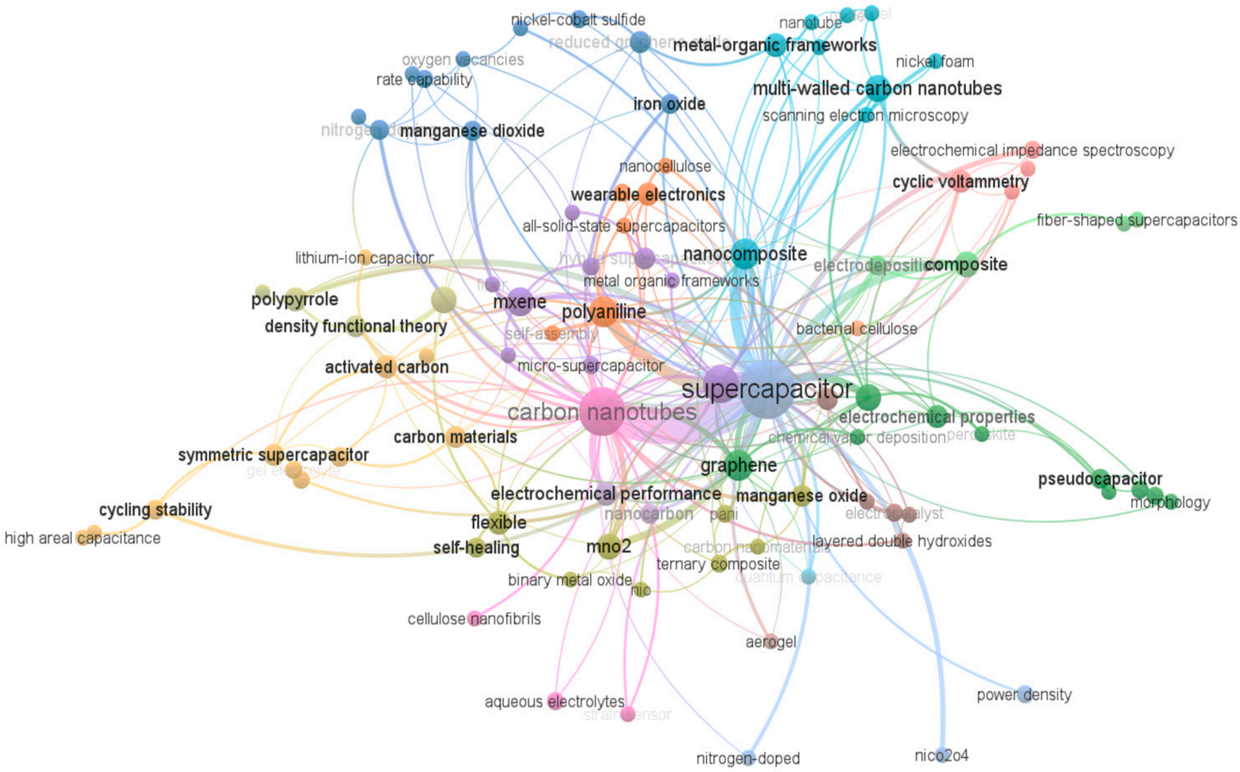

Figure 3. Co-occurrence keywords analysis of CNTs–based supercapacitor by VOSviewer.

Nanomaterials are widely used for industrial applications such as supercapacitors, batteries, antibacterial activity, nano-membranes, and sensors [47–56]. Graphene, due to its highly tunable surface area, outstanding electrical conductivity, good chemical stability, and excellent mechanical behavior, is promising for applications in supercapacitors. The recent development of graphene-based supercapacitors from zero-dimensions to three-dimensions was reported in Ref. [57]. Lu et al. [58] reported a symmetric supercapacitor based on the CNTs/reduced graphene oxide (rGO) electrodes at different temperatures. They demonstrated that the specific capacitances of the symmetric CNT/rGO supercapacitors reached 107.8 and 128.2 F/g at 25 °C and 80 °C, and 80.0 and 144.6 F/g at −20 °C and 60 °C. The CR of CNT/rGO supercapacitors remained almost unchanged after 20,000 cycles. Flexible supercapacitors are used in portable and wearable electronics. The flexible supercapacitors are based on CNTs/graphene, CNTs/GO, and CNTs/SnS_2 compared in Ref. [59]. The paper presented the areal and volumetric capacitance of the symmetric CNT/SnS_2 supercapacitors, which were 533 mF/cm^2 and 63 mF/cm^3, respectively, whereas CNT/GO-based electrodes display a high-rate capability. Zhang et al. [60] investigated the fabrication of the supercapacitor based on the CNT/graphene/FeCo electrode. They stated that the specific capacitance of the CNT/graphene/FeCo supercapacitor was 560.26 F/g and that the supercapacitor exhibits better electron conductivity and electrochemical performance than pure graphene. The electrical properties of supercapacitors based on the CNTs/COOH/Fe_3O_4 and CNT/COOH/Fe_3O_4/rGO structures were compared in Ref. [61]. The paper demonstrated that the specific capacitance, energy, and power densities of the CNT/COOH/Fe_3O_4 and CNT/COOH/Fe_3O_4/rGO supercapacitors were 167.29 F/g, 41.65 Wh/kg, and 1333 W/kg and 448.10 F/g, 34.02 Wh/kg, and 1282 W/kg, respectively. The cyclic stability of the CNT/COOH/Fe_3O_4 supercapacitor was 69% up to 2000 cycles, and for the CNT/COOH/Fe_3O_4/rGO supercapacitor, it had 93% cyclic stability after 2000 cycles. With the explosive growth of portable electronic devices and electric vehicles, the consideration of renewable energy and the development of sustainable green energy storage devices are more highlighted. In another study, a supercapacitor based on the CNT/rGO-aminoindle structure was reported in Ref. [62]. The paper mentioned using redox-active 7-aminoindole (7-Ai) effectively, the agglomeration of rGO nanosheets was reduced, and a rapid diffusion and transport of ions/electrons from network channels constructed by CNTs happened. The specific capacitance of CNT/rGO-aminoindle supercapacitor, power, and energy densities was 183.86 mAh/g, 17.8 kW/kg, and 163.63 Wh/kg, respectively, with a cycling stability of 87.12% for 16,000 cycles. Juang et al. [63] produced graphene sheets via an amino-assisted liquid phase exfoliation method and proposed that the electrical conductivity of functionalized CNT/graphene buckypaper was 87.500 S/m, which is six times higher than unfunctionalized CNT/graphene buckypaper. They stated that for functionalized graphene/CNT buckypaper, the capacitance was 359.6 mF/cm^3 with no capacitance change after 10,000 cycles. A study investigated the preparation of a composite consisting of polypyrrole-derived highly defective CNTs and highly conductive rGO [64]. The highly defective structures serve as effective electrochemically active sites to enhance ion adsorption. Meanwhile, the CNTs and rGO structures act as rapid electrons/ions' dual transport channels. The combination of CNTs and rGO structures with defective structures synergistically promotes fast energy storage/release. The paper demonstrated that the carbon composite with an optimized structure had a specific capacitance of 160.6 F/g. Li et al. [65] fabricated a supercapacitor with the CNT/rGO/PANI structure, and PANI was grown vertically on CNT/rGO fiber. This structure enabled fast transport for electrons and rapid diffusion for ions. For the CNT/rGO/PANI supercapacitor, the volumetric capacitance of 50.2 F/cm^3 with 62.5% CR was achieved A supercapacitor based on the CNT/graphene nanoribbons/molybdenum disulfide (MoS_2) electrode displayed a specific capacitance of 282 F/g [66]. Mendoza et al. [67] compared three types of flexible supercapacitors with structures of CNTs, CNTs/Boron nitride (BN)/graphene, and CNT/Boron nitride (BN)/graphene/lithium titanate (Li_2TiO_3). They reported that the specific capacitance of CNTs, CNTs/BN/graphene, and CNT/BN/graphene/Li_2TiO_3 supercapacitors was 329.7 F/g, 890.2 F/g, and 1662.2 F/g, respectively. On the other hand, the added BN/graphene and BN/graphene/Li_2TiO_3 increased the capacitance by ~4 times. The fabrication of graphene fiber as an electrode for CNT/rGO fiber supercapacitors, using GO/polyacrylonitrile (PAN) fibers, is researched in Ref. [68]. The paper described a specific capacitance increase from 34 F/g to 141 F/g with a CR of 96% after 2500 cycles that was due to the improved inaccessibility and pseudo-capacitance provided by oxygen-containing functional groups. CNT/graphene nanocomposites have drawn scientific attention as effective electrodes for supercapacitors. A supercapacitor based on the CNTs/graphene electrode and 1 M NaCl as the electrolyte showed SC of 179 F/g [69]. The advancement of multifunctional materials and their carbonaceous hybrids as supercapacitor electrodes are of critical importance. Das et al. [70] reported a supercapacitor based on the ternary germanium selenide (Ge_4Se_9) with CNTs and reduced graphene oxide (RGO) and 1 M H_2SO_4 as the electrolyte. They observed that the SC of the supercapacitor was 440 F/g with 98% coulombic efficiency after 5000 cycles. A study investigated a supercapacitor constructed by the tungsten trioxide (WO_3)/CNT/RGO electrode and demonstrated the SC of 691.38 F/g at 5 mV/s [71]. Ngo et al. [72] fabricated polydopamine/CNT/graphene oxide nanocomposites as the flexible symmetric electrode for the supercapacitor and demonstrated the SC of 217.4 F/g. The supercapacitors based on the CNTs/MnO_2 nanocomposite electrode have been developed to fabricate high-performance supercapacitors [73–78]. The dispersion of MnO_2 and conductive additives in an electrode are key factors in increasing the efficiency of the electrode. The high theoretical capacitance of MnO_2 offered it as a promising electrode for supercapacitors. Yesilbag et al. [76] compared the supercapacitors with CNTs and CNT/MnO_2 electrodes and demonstrated that the SC of supercapacitors were 53.7 F/g and 497 F/g, respectively. This review aimed to describe the supercapacitors based on the CNTs, graphene, metals, and polymer electrodes. Furthermore, the SC, energy, and power densities were investigated for CNT-based supercapacitors. The developments of CNT-based supercapacitors for 2022 publications (Scopus) were also compared.

2. CNT/Graphene-Based Supercapacitors

CNTs, due to their unique electrical, mechanical, and chemical stability properties, can be used for the development of supercapacitors, as electrodes create a large interface between the electrode and electrolyte. To obtain a higher energy density, CNTs should be composites with other nanomaterials and conductive polymers [79]. Figure 4 shows the number of publications of CNT/graphene-based supercapacitors in different years. A comparison of the SC of CNT/graphene-based supercapacitors that was published in 2022 is also plotted in Figure 4.

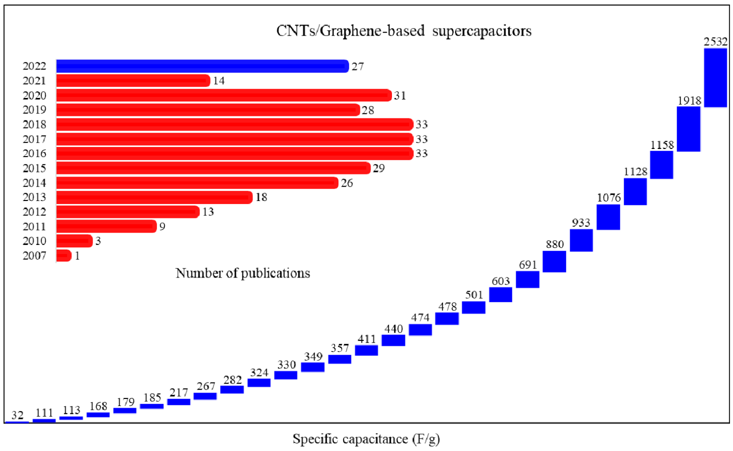

Figure 4. Comparison of SC of CNT/graphene-based supercapacitors as a function of the number of publications.

As can be observed in Figure 4, the SC of different publications in 2022 are from 113 F/g to 2532 F/g. Yang et al. [80] reported a supercapacitor with nanocomposite electrodes based on the CNTs/graphene and Ti_3C_2Tx MXene. They demonstrated that the SC of the supercapacitor at the 3000 mV/s was 349 F/g with a CR of 97.1% after 100,000 cycles. In another study, a supercapacitor based on nanocomposite electrodes with the combination of CNTs, reduced graphene oxide (rGO), and ZrO_2 nanoparticles, was investigated [81]. That study demonstrated that the CNT/rGO/ZrO_2 nanocomposite electrode offers the SC of 357 F/g at 1A/g with a CR of 97.1% after 5000 cycles, and the structure of CNTs/rGO creates an excellent conductive network. Wang et al. [82] described a flexible supercapacitor with a CNT/rGO electrode and demonstrated a SC of 267.8 F/g. Recently, paper supercapacitors based on the carbon nanomaterials' electrode and polymer electrolyte were used. Figure 5 shows a schematic of paper-based supercapacitors.

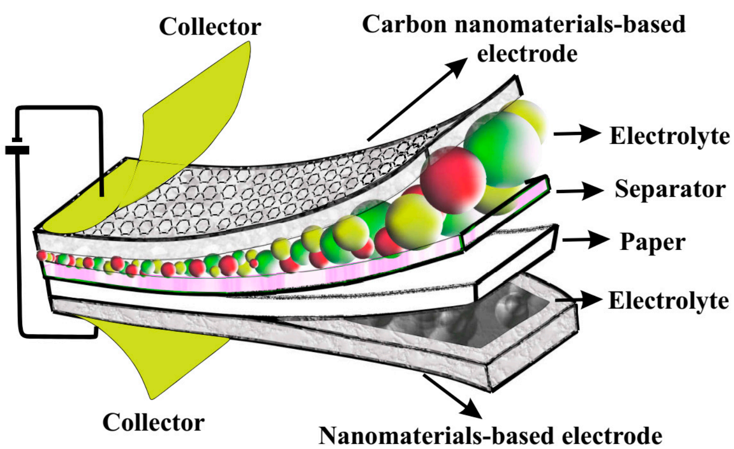

Figure 5. Schematic of typical symmetry paper-based supercapacitor.

The comparison of CNT/graphene electrode-based supercapacitors is shown in Table 1. Liu et al. [83] described a hybrid supercapacitor with polyhedron-CNTs/graphene as the negative electrode and CNT/$NiCo_2O_4$/CoP core-sell polyhedron as the positive electrode. The study demonstrated that the polyhedron-CNT film with porous structure and high conductivity achieved an SC of 1918.4 F/g, and energy and power densities of 68.6 Wh/kg and 800 W/kg at 1 A/g, respectively. The asymmetric supercapacitor based on the CNT/rGO/FeO/NiFe electrode was investigated in Ref. [84]. They reported that the SC, and energy and power densities of the supercapacitor were 411.9 F/g, 41.4 Wh/kg, and 5600 W/kg, respectively, at a scan rate of 5 mV/s and CR of 102.2% after 5000 cycles.

Table 1. Comparison of CNT/graphene electrode-based supercapacitors.

Electrode Materials	CR (%)/Cycles	SC (F/g)	Energy Density (Wh/kg)	Power Density (W/kg)	Reference
CNTs/graphene/polyhedron	-	1918.4	68.6	800	[83]
CNTs/rGO/FeO/NiFe	102.2/5000	411.9	41.4	5600	[84]
CNTs-GONRs@g-C3N4/Ni-Co-LDH/NF	-	2532.80	77.61	850	[85]
CNTs/graphene/$NiCo_2O_4$/$ZnCo_2O_4$	86.1/6000	1128.6	68.6	800	[86]
CNTs/graphene/N-Co_3S_4	97.2/4000	1158	37.69	8000	[87]
CNTs/graphene/N-MnO_2	-	185.1	75.3	18,100	[88]
CNTs/graphene/$NiCo_2S_4$/MXene	80/5000	1076	0.115 Wh/m^2	12.4 W/m^2	[89]
CNTs/graphene/Mn0.06CO_2.94O_4	82/33,000	933	55	9000	[90]
CNTs/graphene	-	603	0.24 Wh/m^2	21.6 W/m^2	[91]
CNTs/graphene/activated carbon	94–97/1000	32.13	6.6	69	[92]
CNTs/graphene/TiO_2	-	168	15	337.5	[93]

Qiu et al. [85] used nanocomposites based on CNTs, GO/nanoribbon@graphitic, carbon nitride/Ni and Co-layered double hydroxide/Ni foam (CNTs-GONRs@g-C_3N_4/Ni–Co-LDH/NF) for the supercapacitor electrode. They proposed that the SC of the supercapacitor at 1A/g was 2532.80 F/g and energy and power densities were 77.61 Wh/kg and 850 W/kg, respectively. Chen et al. [86] considered a nanocomposite based on CNTs/graphene/$NiCo_2O_4$/$ZnCo_2O_4$ as the electrode for an energy storage device. They indicated that the SC of the supercapacitor was 1128.6 F/g at 1A/g with a CR of 86.1% after 6000 cycles at 10A/g. That nanocomposite demonstrated an energy density of 68.6 Wh/kg and a power density of 800 W/kg at 1 A/g. Transition metal sulfides due to high theoretical capacity have become electrodes in supercapacitors. Zhang et al. [87] proposed a supercapacitor electrode based on the CNTs/graphene/N-Co_3S_4. They demonstrated that the CNT/graphene/N-doped -Co3S4 supercapacitor has energy and power densities of 37.69 Wh/kg and 8000 W/kg, respectively. In another study, the supercapacitor based on the CNT/graphene/N-doped-MnO_2 electrode exhibited energy and power densities of 75.3 Wh/kg and 18,100 W/kg, respectively [88]. Hybrid nanostructures have been used to design flexible supercapacitors. However, the performance and low cost of supercapacitors are interesting characteristics. Pathak et al. [89] reported the supercapacitor based on the MXene (Ti_3C_2Tx) as the negative electrode and CNT/graphene/$NiCo_2S_4$/MXene nanocomposites as the positive electrode. They proposed that the SC of the supercapacitor was 1076 F/g. Transition metal oxides, CNTs, and graphene have been composited as possible electrodes for high-performance supercapacitors. The supercapacitor with CNT/graphene/$Mn_{0.06}Co_{2.94}O_4$ structure displayed a high SC of 933 F/g [90]. The process for preparing ionic gel and CO_2 plasma treatment to modify electrodes based on CNT/graphene composites is presented in Ref [91]. The paper demonstrated that the SC of the supercapacitor was 603 F/g, and the energy and power densities of the supercapacitor were 24.03 µWh/c and 2.16 mW/cm, respectively. Rustamaji et al. [92] designed and fabricated a supercapacitor based on the AC/CNT/graphene electrode. They reported that the SC of the AC/CNT/graphene supercapacitor was 32.13 F/g, and the power and energy densities of the supercapacitor were 69 W/kg and 6.6 Wh/kg, respectively. The supercapacitor based on the CNT/rGO/TiO_2 electrode demonstrated the SC of 168 F/g and energy and power densities of 15 Wh/kg and 337.5 W/kg, respectively [93].

3. CNT/PANI-Based Supercapacitors

Polymers due to electrochemical properties were used in both supercapacitors' electrodes and electrolytes [94–96]. Conducting polymers such as polyaniline (PANI) have recently been widely used in the electrode material composite of supercapacitors for the fabrication of a lightweight, flexible, symmetry, and high-performance energy storage device. Panasenko et al. [97] reported the supercapacitor based on the CNT/PANI electrode with a SC of 541 F/g. The supercapacitor is based on the CNT/PANI/rGO electrode with a SC of 478 F/g presented in Ref. [98]. The micro-supercapacitors with CNT/PANI/MXene electrodes showed the SC of 414 F/g and cycling stability of 90.4% after 10,000 cycles [99]. Electrode material based on the PANI composite is ideal for flexible supercapacitors. However, the formation of PANI agglomeration leads to a decrease in the SC and cycling stability. The supercapacitor with the CNT/PANI/Sb composite electrode demonstrated the SC of 416 F/g [100]. A comparison of the SC, power, and energy densities of CNT/PANI-based supercapacitors published in 2022 is mentioned in Table 2. In Ref. [101], the supercapacitor based on the CNT/PANI/MnO_2 electrodes is developed for energy storage applications and exhibited a high SC of 532 F/g at a current density of 1 A/g. Moreover, the paper demonstrated 80% CR after 4000 cycles and had energy and power densities of 11.8 Wh/kg and 3785 W/kg, respectively. MXene, a successful nanomaterial with a two-dimensional structure, has displayed a capable application on energy storage devices. Due to the susceptibility of MXene to oxidation as a cathode electrode, there is a need for research into developing these electrodes. Wu et al. [102] introduced a supercapacitor with a CNT/PANI@MXene electrode and reported the SC of 463 F/g at 5 mV s−1, and 92% CR after 10,000 cycles. They mentioned the supercapacitor with the composite cathode, and the MXene/CNTs anode achieved energy and power densities of 10 Wh/kg and 2808 W/kg, respectively. In a study, CNTs were embedded in polydimethylsiloxane for outstanding adhesion and integration and then PANI was deposited on its surface [103]. The study proposed that for the CNT/PANI/Polydimethylsiloxane composite electrode, the SC, power, and energy densities of the supercapacitor were 265 F/g, 25.5 Wh/kg, and 126.6 W/kg, respectively. In composite electrodes (nanomaterials/PANI), the electrochemical enhancement of PANI is related to the dispersion state of the nanomaterials in the polymer. Wang et al. [104] demonstrated the fabrication of a symmetric supercapacitor based on the CNT-lignosulfonate/PANI/molybdenum disulfide-lignosulfonate (CNTs-LS/MoS_2-LS/PANI) electrode. They demonstrated that, in comparison to the supercapacitor based on the pure PANI, the performance of the supercapacitor with nanocomposites' electrode enhanced. They reported that the SC, power, and energy densities of the supercapacitor with the CNT-LS/MoS_2-LS/PANI nanocomposite electrode were 458.9 F/g, 265.3 W/kg, and 10.9 Wh/kg with 86% CR after 10,000 cycles. Fabrication of a symmetrical supercapacitor was based on the CNT/PANI/graphene/graphite electrode with a high surface area, described in Ref. [105]. The paper demonstrated the SC of 880 F/g at a current density of 1.5 A/g, an energy density of 68.4 mWh/g, a power density of 895.4 mW/g, and 80.6% CR after 10,000 cycles. Xu et al. [106] demonstrated a flexible, symmetric supercapacitor based on the CNT/PANI-Carboxymethylcellulose (CMC) electrode with a gravimetric SC of 348.8 F/g with 89.2% CR after 5000 cycles. Furthermore, they exhibited that the energy and power densities of symmetric supercapacitors with the CMC-PANI/CNT electrode were 99.89 μW h/cm^2 and 400.02 μW/cm^2, respectively. The supercapacitor with the CNT/vinyltrimethoxysilane/PANI flexible electrode was introduced in Ref. [107] and displayed the highest SC and energy density of 531.3F/g and 26.5 Wh/kg, respectively, at the current density of 1 A/g.

Table 2. Comparison of CNT/PANI electrode-based supercapacitors.

Electrode Materials	CR (%)/Cycles	SC (F/g)	Energy Density (Wh/kg)	Power Density (W/kg)	Reference
CNTs/PANI/MnO_2	80/4000	532	11.8	3785	[101]
CNTs/PANI@MXene	92/10,000	463	10	2808	[102]
CNTs/PANI/PDMS	76/5000	265	25.5	126.6	[103]
CNTs-LS/MoS2-LS/PANI	86/10,000	458.9	10.9	265.3	[104]
CNTs/PANI/graphene/graphite	80.6/10,000	880	68.4	895.4	[105]
CNTs/PANI-Carboxymethylcellulose	89.2/5000	348.8	0.999 Wh/m^2	4 W/m^2	[106]
CNTs@vinyltrimethoxysilane/PANI	96.6/2000	531.3	26.5	-	[107]

4. CNTs/Metal-Based Supercapacitors

Increasing energy demands are causing the growth of new materials for supercapacitors' electrodes and electrolytes. Recently, CNT/metal nanocomposites were widely investigated for the development of supercapacitor electrode materials such as CNTs/HCNFs/MOF, CNTs/$NiPMo_{12}$, CNT/BT/Zn-N, CNTs@NHP/Co@LDH-CoZnAl, CNTs/$ZnCo_2O_4$, CNTs/NiO/MnO_2, CNTs/Ti_3C_2TX, CNTs/KCl, CNTs/V_2CTx MXene, CNTs/$NiC_{32}N_8H_{16}$, CNTs/MnS, CNTs/MnO_2, CNTs/Ni–Va and CNTs/$ZnWO_4$/Ni foam, CNTs/NiO/MnO_2, CNTs/ZnO/C, CNTs/$NiCo_2S_4$@N, CNTs@Bi_2S_3/MoS_2, CNTs/$Cu_2P_2O_7$, and CNTs/MXene with a SC of 712 F/g, 815.6 F/g, 252 F/g, 869.6 F/g, 888 F/g, 23 F/g, 270.5 F/g, 334.4 F/g, 1842 F/g, 12 F/g, 8 F/g, 1298 F/g, 386 F/g, 1493 F/g, 4552 F/g, 1320 F/g, 650 F/g, 254 F/g, 1338 F/g, 465 F/g, and 401.4 F/g, respectively [108–127]. A comparison of SC, power, and energy densities of CNT/metal-based supercapacitors published in 2022 is mentioned in Table 3.

Table 3. Comparison of CNT/metal electrode-based supercapacitors.

Electrode Materials	CR (%)/Cycles	SC (F/g)	Energy Density (Wh/kg)	Power Density (W/kg)	Reference
CNTs/Ni	83.2/5000	97	32.6	476.5	[128]
CNTs/$NiCo_2O_4$/Ni/C	-	1945.9	58.31	749.7	[129]
CNTs/$NiCoO_2$	92/5000	1587	41.8	412	[130]
CNTs/NiS_2/$NiCo_2S_4$	83/10,000	2905	58.1	-	[131]
CNTs/$NiFe_2O_4$	89.16/5000	670	23.39	466.66	[132]
CNTs/Co-S@carbon nanofibers	96.9/10,000	416.5	10.3	320	[133]
CNTs/$Co_3V_2O_8$	95.26/3000	120.17	37.55	-	[134]
CNTs/ZIF/MoS_2	-/10,000	262	52.4	3680	[135]
CNTs/ZnCoS	96/10,000	2957.6	68.8	700	[136]
CNTs/MnO_2	78.26/6000	253.86	32	413.70	[137]
CNTs/Ti_3C_2/$MnCo_2S_4$	94.09/5000	823	49.5	350	[138]
CNTs/Bi-Fe-P	85.6/8000	532	81.5	890.2	[139]
CNTs/cerium selenide nanopebbles	84.1/4000	451.4	36.3–14.5	2800–5600	[140]
CNTs/Nb_2O_5	96/10,000	192	5.4–2.7	98.7–24.671	[141]
CNTs/Nitrogen–Boron–Carbon	95.7/3000	432.31	11.67–7.74	300–1485	[142]
CNTs/SnS_2-BN	101/-	87	49	-	[143]

Metals that are organic due to chemical and physical properties such as porosity were proposed for supercapacitors' electrode material. A comparison of supercapacitors based on CNT/Ni electrode structures is reported in Table 3 [128–132]. Sun et al. [128] synthesized the CNT/Ni composite electrode for the supercapacitor electrode and reported that the SC of the supercapacitor was 97 F/g and 83.2% retention after 5000 cycles. They reached 32.6 Wh/kg and 476.5 W/kg energy density and power density, respectively, for CNT/Ni supercapacitors. The hierarchical electrode materials are attractive for energy storage applications. The supercapacitor is based on the CNT/$NiCo_2O_4$/Ni/C electrode structure described in Ref. [129]. The paper mentioned that the SC of a supercapacitor with the hierarchical electrode material was 1945.9 F/g with an energy density of 58.31 Wh/kg at a power density of 749.7 W/kg. One of the disadvantages of supercapacitors is related to the low energy density that can be improved by pseudocapacitive materials from redox reactions. A supercapacitor electrode based on the CNT/$NiCoO_2$ mesoporous structure presented the SC of 1587 F/g and 92% CR after 5000 cycles, with an energy density of 41.8 Wh/kg at a power density of 412 W/kg [130]. Geioushy et al. [131] introduced the CNT/NiS_2/$NiCo_2S_4$ nanostructure as electrode material for energy storage applications. They achieved the SC of 1587 F/g and 83% CR after 10,000 cycles. The composition of the binary metal oxide with CNTs is considered for increased energy storage in recent reports. The supercapacitor with the CNT/$NiFe_2O_4$ electrode structure demonstrated energy and power densities of 23.39 Wh/kg and 466.66 W/kg, respectively [132]. Supercapacitors with long-cycle stability and flexible electrodes are in demand for portable electronic equipment. Yao et al. [133] investigated the flexible quasi-solid-state supercapacitor based on the CNT/Co-S/carbon nanofibers' electrode and stated that the SC of the supercapacitor was 416.5 F/g and 96.9% CR after 10,000 cycles. Due to the tunable structure and stability properties of the binary metal, oxides were used for electrode materials' construction. The supercapacitor with the gel polymer electrolyte and the CNT/$Co_3V_2O_8$ electrode was proposed in Ref. [134]. The paper detailed that the SC of the supercapacitor was 120.17 F/g and 95.26% CR after 3000 cycles. Despite the capability of supercapacitors in energy storage, there is a big difference in energy densities between batteries and supercapacitors. Houpt et al. [135] introduced a hybrid framework with the CNT/ZIF/MoS_2 electrode material. They stated that the power density was 3682 W/kg for the supercapacitor with a SC of 262 F/g. A supercapacitor based on the metal–organic CNT/ZnCoS electrode demonstrated the SC of 2957.6 F/g with a 96% CR after 10,000 cycles [136]. Metal–oxygen material such as CNTs/MnO_2 was used for the supercapacitor electrode and achieved the SC of 253.86 F/g with 78.26% CR after 6000 cycles, with energy and power densities of 32 Wh/kg and 413.7 W/kg, respectively [137]. The improvement of supercapacitors' performance is related to the electrode materials and structural design. The CNT/Ti_3C_2/$MnCo_2S_4$ composite electrode with positively charged CNTs and negatively charged Ti_3C_2 was investigated in Ref. [138]. The paper mentioned that the symmetric supercapacitor demonstrated the SC of 823 F/g and energy and power densities of 49.5 Wh/kg and 350 W/kg respectively. Pseudocapacitive materials due to their good redox processes and low cost are proper for supercapacitor fabrication. A supercapacitor based on the CNT/Bi-Fe-P electrode exhibited an energy density and power densities of 81.5 Wh/kg and 890.2 W/kg, respectively, with 85.6% CR after 8000 cycles [139]. Problems in flexible energy storage devices are related to mechanical properties, cyclic stability, and small capacitance. A flexible symmetric supercapacitor based on the CNT/cerium selenide nanopebbles' electrode and poly (vinyl alcohol) (PVA)-$LiClO_4$ gel electrolyte was proposed in Ref. [140]. The study achieved energy and power densities in a range of 36.3–14.5 Wh/kg and 2800–5600 W/kg, respectively. There is sodium due to an abundance in the Earth's crust, leading to low-cost energy storage and renewable energies. In combination with niobium pentoxide and CNTs, sodium ions can make intercalation into niobium pentoxide and electrostatic adsorption into CNTs.

Real et al. [141] investigated a flexible, sodium-ion pseudocapacitor with the CNT/Nb_2O_5 electrode and demonstrated a SC of 192 F/g and 96% CR after 10,000 cycles. A supercapacitor based on the CNT/Nitrogen–Boron–Carbon electrode reached energy density and power density in a range between 11.67–7.74 Wh/kg and 300–1485 W/kg, respectively [142]. Due to large-volume oscillations, the low inherent conductivity and stability of the bare SnS2 are not expected for supercapacitor devices. A supercapacitor with CNT/SnS_2-BN electrode material stated the SC of 87 F/g [143]. A comparison of Tables 1–3 in the specific capacitance, power density, and energy density for supercapacitors based on the CNTs/metal, CNTs/graphene, and CNT/PANI is plotted in Figure 6. As can be observed from Figure 6 for CNT/metal-based supercapacitors, the energy density is higher, and for CNT/graphene-based supercapacitors, the power density is highlighted. The

CNT/metal-based supercapacitors achieved an energy density of about 80 Wh/kg, and the CNT/graphene-based supercapacitor reached a power density of 9000 W/kg.

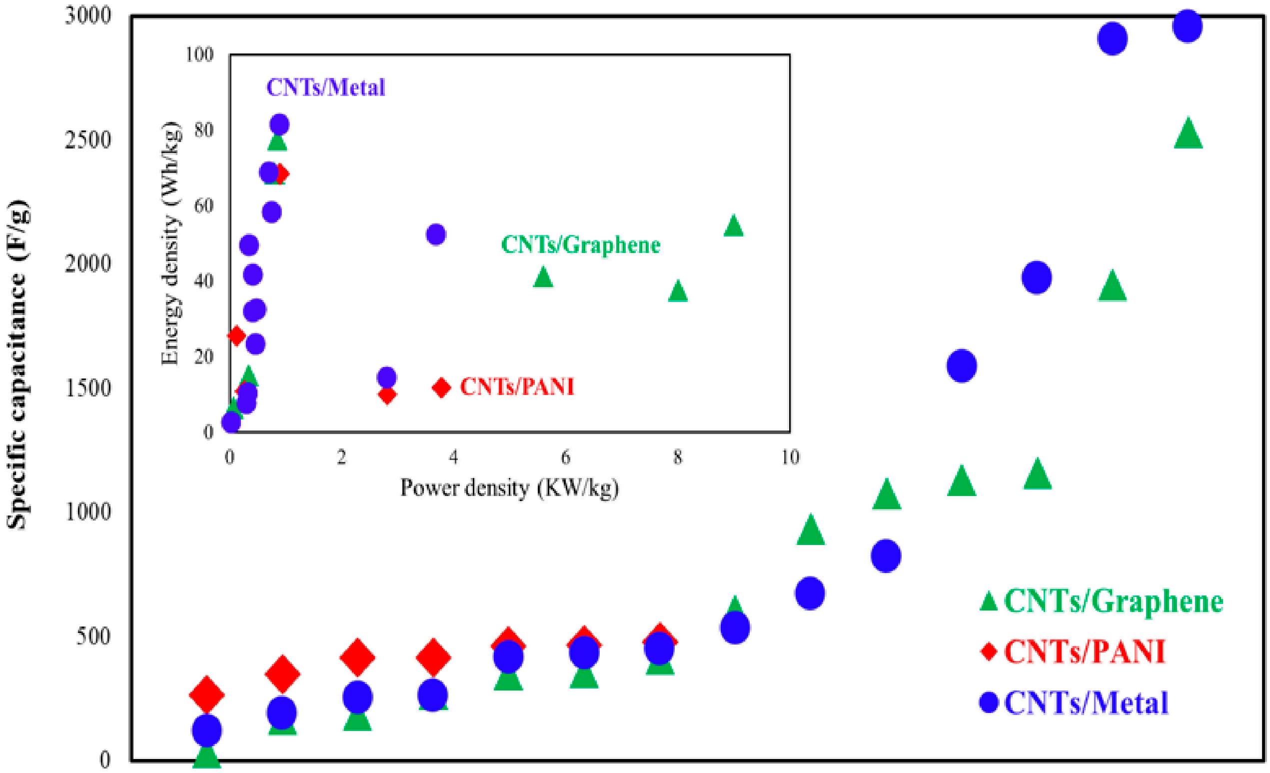

Figure 6. Comparison of specific capacitance, power density, and energy density for supercapacitors based on the CNTs/metal, CNTs/graphene, and CNTS/PANI.

5. Conclusions

There is a continued need for supercapacitors in several industries for energy storage systems due to fast charge/discharge, high cycle stability, and high-power density. In this study, a recent development in CNT-based supercapacitors was reported. The comparison of supercapacitors based on the CNTs/graphene, CNTs/metal, and CNTs/polymer electrodes was investigated in this review. Results demonstrated that for the CNT/metal-based supercapacitors, the energy density is higher and for CNT/graphene-based supercapacitors, the power density is highlighted. The CNT/metal-based supercapacitors achieved an energy density of about 80 Wh/kg, and the CNT/graphene-based supercapacitor reached a power density of 9 kW/kg.

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