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Review

Hybrid Nanostructured Materials as Electrodes in Energy Storage Devices

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Abstract: The global demand for energy is constantly rising, and thus far, remarkable efforts have been put into developing high-performance energy storage devices using nanoscale designs and hybrid approaches. Hybrid nanostructured materials composed of transition metal oxides/hydroxides, metal chalcogenides, metal carbides, metal–organic frameworks, carbonaceous compounds and polymer-based porous materials have been used as electrodes for designing energy storage systems such as batteries, supercapacitors (SCs), and so on. Different kinds of hybrid materials have been shown to be ideal electrode materials for the development of efficient energy storage devices, due to their porous structures, high surface area, high electrical conductivity, charge accommodation capacity, and tunable electronic structures. These hybrid materials can be synthesized following various synthetic strategies, including intercalative hybridization, core–shell architecture, surface anchoring, and defect control, among others. In this study, we discuss applications of the various advanced hybrid nanostructured materials to design efficient batteries and SC-based energy storage systems. Moreover, we focus on their features, limitations, and real-time resolutions.

Keywords: hybrid materials; energy storage systems; battery; supercapacitor; hybrid electrode

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1. Introduction

The radical deficit of fuel, global warming, and the growing interest in moveable electronic devices and electric automobiles have encouraged the development of efficient energy storage systems. Moreover, the total energy consumption rate is exponentially growing with the increasing global population, economic revolutions, invention of technologies and machines, access to modern facilities in remote areas, and significant changes to human lifestyles [1]. According to the annual BP Statistical Review of World Energy 2022, the total amount of primary energies consumed worldwide in 2021 exceeded 595 exajoules, which is 3% higher than that in 2019 and 5.5% higher than in 2020, when primary energy consumption levels fell due to the coronavirus pandemic and its effects on the demand for transportation, fuel, and general economic performance (Figure 1) [2]. Another report has predicted that the demand for clean and sustainable energy will increase by about 20% by 2050 [3]. The radical shortage of fossil fuels and growing interest in wearable electronics and electric vehicles has driven the innovation of efficient hybrid technologies for suitable energy storage systems.

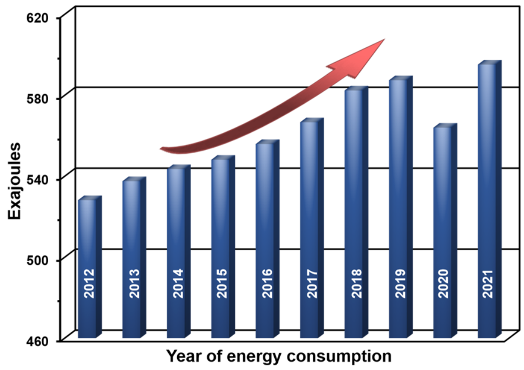

Figure 1. Statistics of the world's primary energy consumption for the last 10 years.

In order to maintain a balance between energy production and consumption, it is essential to store excess energy to fulfil the demand in relation to short- and long-term purposes. The utilization of advanced energy storage facilities makes it possible to overcome the limitations of suitable storage systems for renewable energies from solar, wind, hydroelectric, and geothermal sources. In this case, non-conventional energy storage devices—predominantly batteries and capacitors—facilitate the storage of electricity from renewable sources. Moreover, it is possible to distribute it continuously through electric wires, even in remote areas. Therefore, there have been significant research efforts focused on the innovation of sustainable energy storage technologies. Among them, priority has been given to rechargeable batteries and capacitors due to their remarkable storage capacity, with very high power (W kg^{-1}) or energy density (W h kg^{-1}) [4].

Developing new devices to satisfy the necessity and urgency of energy storage has become an important issue for addressing the challenges regarding energy supply. The International Energy Agency (IEA) reports that worldwide energy demand will increase up to 450 GW in 2050 [5]. Therefore, it is important to store the excess energy for future use, if any extra electricity is produced from any source. For a long time, batteries have been used as non-conventional devices for storing and supplying energy in remote areas. Many different types of vehicles are run using the energy stored in batteries as a portable form. The latest applications of this type of energy are in smartphones (e.g., Galaxy and iPhones); in this case, lithium-ion (Li-ion) batteries play a vital role, storing and supplying energy for operating these electronic devices. High-tech manufacturing facilities with continuous power supply at a consistent frequency are essential for the production and use of such electronic devices [6]. Compared to the elevated rate of energy demand, a slow transition rate has been observed in developing sustainable energy systems. Currently, the biggest challenge in satisfying the rising energy demand is introducing an efficient and cost-effective energy storage system. Additionally, it is important to develop efficient materials for manufacturing devices with high capacity, such as supercapacitors and rechargeable batteries. Therefore, the biggest difficulty facing electrochemical systems in recent years has been the investigation of new electrode materials or the modification of existing ones. In particular, the specific capacity, cyclic stability, and Columbian efficiency are three very important properties desired for any energy storage cell to work effectively. However, there are common limitations of most materials used as electrodes, such as limited capacity, changes in crystal structure, and volume growth during cycling [7,8].

To avoid these drawbacks, hybrid nanostructured materials are being employed as electrode materials in modern supercapacitors as well as various batteries. Hybrid materials are nanocomposite materials with or without interactions between the inorganic and organic components, amorphous sol-gel complexes, and crystalline highly structured coordination polymers, etc. Therefore, to increase the power and energy density of next-generation storage systems, varieties of multifunctional hybrid nanostructured materials are consistently generating tremendous interest.

There are five sections in this review. The various types of energy storage systems, especially supercapacitors and batteries will be concisely covered in Section 2 after the introduction in Section 1. Section 3 will detail the different types of nanostructured hybrid materials, their special electrochemical characteristics, and some representative outcomes as an example of recent development for energy storage performances. The current electrochemical data on hybrid energy storage systems will be briefly examined in Section 4, along with other information. We will also summarize our discussion with some limitations and a forecast for the future applications of hybrid electrode materials in Section 5.

2. Energy Storage Devices

Available energy storage devices can be classified into various types; for example, based on storage duration, there are three types: short-, mid-, and long-term. Depending on their reaction time, they are considered to be either rapid or slow. According to their storage capacity, they can be classified as small-, medium-, or large-scale [9]. There are many techniques for the storage of various types of energies, including electrical, mechanical, chemical, and thermal. Moreover, based on the precise needs and applications, storage technologies have different technical and economic criteria [10].

2.1. Types of Energy Storage Systems

The classification of energy storage systems is consistent with the various associated shapes; energy is often stored within a mold, which can be electrochemical, electromagnetic, mechanical, and thermal one (Figure 2). Hydrogen batteries and fuel cells are electrochemical systems, while electromagnetic systems include SCs and superconductors. Mechanical systems are often divided into kinetic energy storage (e.g., flywheels) and potential energy storage (e.g., pumped hydraulic and compressed gas systems) [11,12].

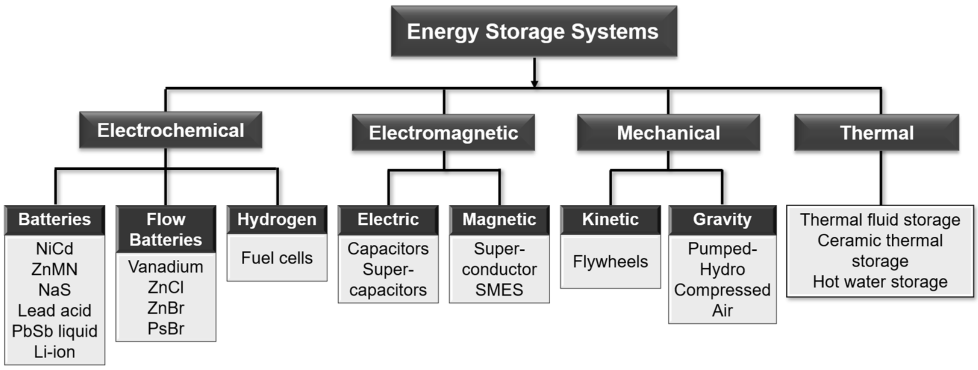

Figure 2. Classification of energy storage systems based on their natural construction.

Techniques and devices are constantly changing and improving to meet the increasing demands for energy storage devices. The battery, as the primary systematic energy memory device, remains the leading and most widely used technique, with a general efficiency of over 90%. A battery may be a chemical device, storing electricity within chemicals and converting the stored energy into a direct current (DC) through an electrochemical reaction. A battery usually consists of three main components: two electrodes and an electrolyte. It also consists of terminals, a separator, and a container. There are two types of electrodes: anode (negative electrode) and cathode (positive electrode) [13]. The Italian physicist Alessandro Volta invented the first battery (in the modern sense) in 1800, which consisted of disks of zinc and copper, while a concentrated salt solution (brine) was used as an electrolyte. To date, many different types of batteries have been reported, such as Ni-Cd, Li-O_2, and Li-S batteries [14,15].

2.2. Batteries for Energy Storage

There are two main types of batteries: physical and chemical batteries (Figure 3a). A physical battery is a device that directly converts solar power, thermal energy, or atomic energy into DC electricity using physical effects, such as solar cells, thermoelectric generators, core batteries, and so on. A chemical battery is a device that converts energy directly into DC electricity. Chemical batteries are the most common type of batteries, and can be classified into two categories: primary (non-rechargeable) and secondary (rechargeable). A primary battery is a straightforward and convenient power source, such as a zinc–manganese or alkaline–manganese battery, which cannot be charged electrically. They are utilized in household applications for various portable electronic and electrical appliances, such as lamps, cameras, watches, toys, radios, and so on. A secondary battery is commonly referred to as a rechargeable battery, which may be recharged after being discharged to its original state by a current through the cells during discharge. For instance, lead–acid and lithium-ion batteries are well-reported rechargeable batteries. It should be noted that the energy storage capacity of an electrochemical system is restricted by the electrochemical properties of the electrode materials. Therefore, storage abilities need to be increased through the use of coupling (or hybridizing) with materials having very low equivalent weights. At present, the world is entering a new era of digital technologies featuring updated energy storage devices.

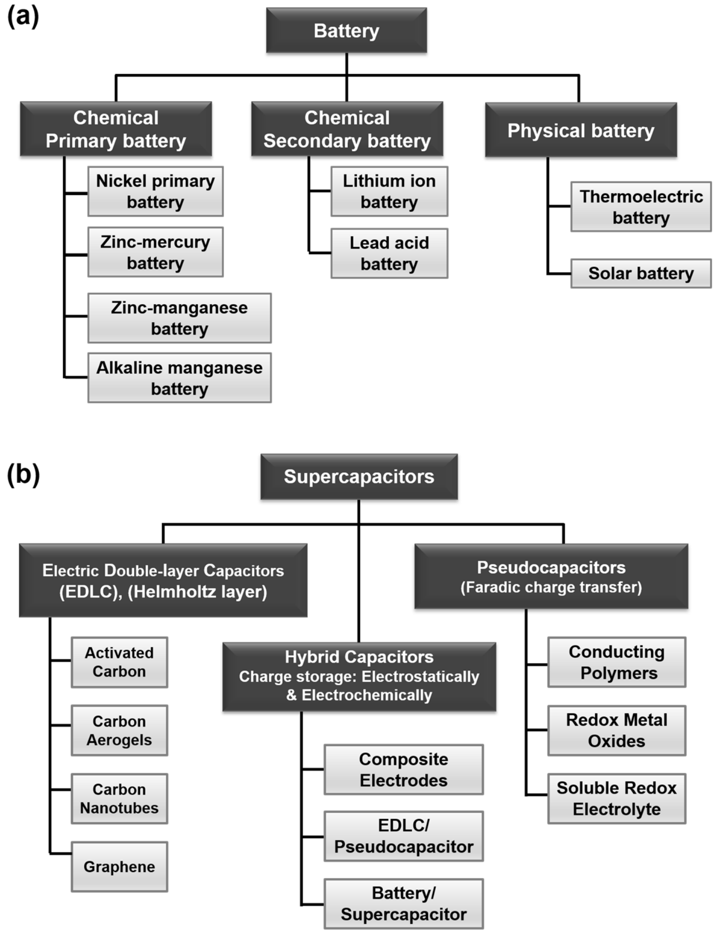

Figure 3. Classifications of: (**a**) batteries; and (**b**) supercapacitors based on electrode materials and charge storage mechanism.

2.3. Supercapacitors for Energy Storage

After the invention of rechargeable batteries, another storage device, known as a capacitor, has been widely used for this purpose. A supercapacitor (SC) is an upgraded version of a capacitor. In this line, the hybridization of a battery and capacitor can be accomplished to achieve high energy storage efficiency. The general performance of an SC is mainly governed by the nature and structure of the electrodes, separator, current collector, and electrolytes. SCs provide many advantages over batteries, such as a high power density (>10 kW kg^{-1}), quicker charging/discharging speed (within seconds), cyclic stability (>100,000 cycles), and being more cost-effective and safer [16,17].

Figure 3b exhibits the different types of SCs. Based on their charge storage behavior and electrode materials, there are three kinds of supercapacitors: electrochemical double-layer capacitors (EDLCs), pseudo-capacitors, and hybrid capacitors [18]. The most well-known type of energy storage technology now employed in industrial applications is the EDLC. Here, electricity is stored at the interface between the electrolyte and electrode through Helmholtz double layers [19]. As this process does not allow for a Faradaic redox reaction, the swelling of the active material, EDLCs typically present high stability and rapid charge/discharge rates. Due to their environmental friendliness, large specific surface area, good electrical conductivity, high chemical stability, and wide working temperature range, carbon-based materials such as graphene, activated charcoal, CNTs are used to make the majority of EDLC electrodes. Furthermore, depending on the type of electrolyte utilized, EDLC performance might be altered. However, the energy density of EDLC equipment is constrained as a result of the electrostatic surface charging mechanism, which severely limits the use of EDLCs.

On the other hand, pseudocapacitor electrodes are constructed of some redox components, such as polymers, metal oxides, or hydroxides [20]. Moreover, their charge storage mechanism follows a fast Faradaic reaction. The formation of such an electrical double layer near the surface provides larger capacitance with high energy density, compared to other capacitors. As pseudocapacitors can store charge through electroporation, redox reactions, or intercalation, they have higher capacitance and energy density than EDLCs. In contrast to EDLCs, pseudocapacitance can be created both on the electrode surface and throughout the entire electrode, resulting in a higher energy density and larger capacitance.

Another type is called hybrid capacitors, which are a combination of EDLCs and pseudocapacitors, with higher specific capacitance compared to each individual type. A hybrid capacitor accumulates charges either electrostatically or electrochemically, using electrochemical or electrostatic absorption-desorption or oxidation-reduction reactions. The hybrid supercapacitor is further divided into three categories: battery-type hybrid, composite hybrid, and asymmetric hybrid. With its extensive power and energy density, as well as improved cycling stability, the hybrid supercapacitor can perform well [21]. In addition, SCs may have various geometric properties, such as flexible SCs including a skinny film, sandwich-type, and planar structure (e.g., as integrated micro-SCs). Flexible SCs are ultrafast rechargeable energy storage devices. It is necessary to utilize a material with very good conductivity, high mechanical rigidity, compact structure, and lightweight properties for the design of flexible SC devices. A planar structure design enhances the rapid ion transfer within the 2D-direction of flexible SCs [22–24].

In terms of the storage and release of electricity, batteries and capacitors are very similar; however, they perform in a completely different manner. In general, batteries offer better energy density for storage, whereas capacitors may charge and discharge more quickly [25]. The supercapacitor's cyclic voltammetry (CV) curve (Figure 4a) is rectangular during the charge and discharge operation, yet the current is nearly constant. Additionally, the galvanostatic charge/discharge (GCD) curve (Figure 4c) of this device is typically inclined with a constant slope. Generally, a battery keeps its voltage steady except when it is close to 100% charged/discharged (TOC/EOD), during which it exhibits Faradaic reactions and its CV curve displays a clear redox peak (Figure 4b,d). In addition, the GCD curve shows a relatively flat charge-discharge platform. At the same time, supercapacitors must be integrated with a DC-DC converter for applications requiring a consistent output voltage to control and maintain the output voltage. The energy stored in these two types of electrodes is measured differently (in terms of capacitance vs. capacity) due to the difference in the charge-storage process [26].

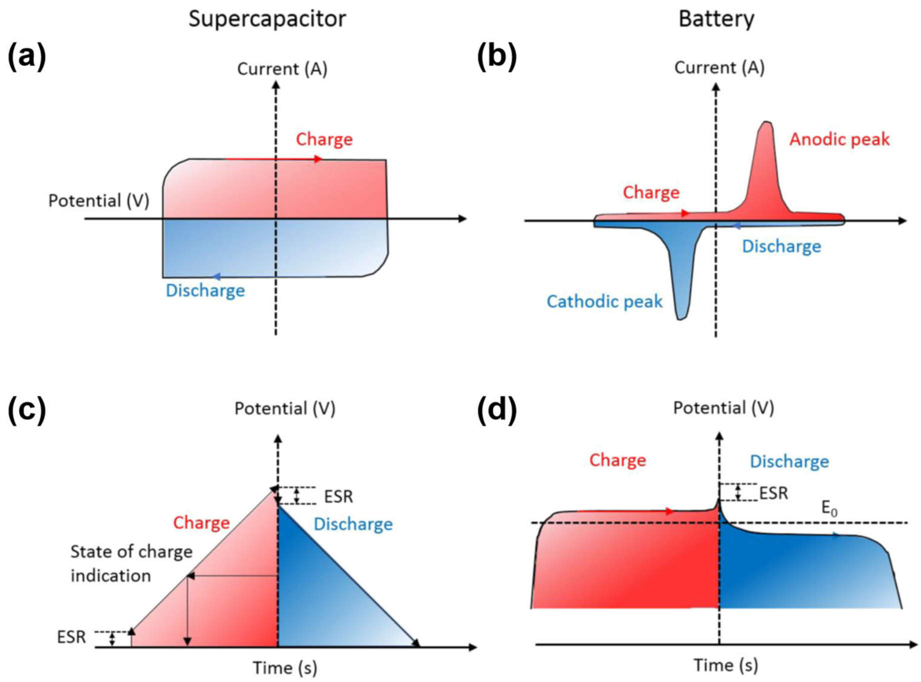

Figure 4. (**a**,**b**) Cyclic voltammetry (CV) curves and (**c**,**d**) galvanostatic charge-discharge (GCD) curves of supercapacitors and batteries. Reproduced with permission from [26]. Copyright 2018 American Chemical Society.

Based on the power density equation, the energy density of a capacitor is determined by the specific capacitance of the electrode material and the potential difference between the positive and negative electrodes, developing porous nanoelectrode materials is one of the best strategies to raise the energy density of supercapacitors. Porous nanoelectrode materials can boost energy density by increasing the specific surface area, which in turn raises the specific capacitance. Constructing hybrid or asymmetric supercapacitors is a further approach that can improve the performance of the entire device. Hence, we highlight current developments in nanostructured hybrid electrode materials for batteries and supercapacitors in the next section.

3. Hybrid Nanostructured Materials in Energy Storage Devices

Hybrid nanostructured materials are a type of nanocomposites made up of two or more separate components at the molecular level, each of which has at least one dimension on the nanoscale. They are put together in a manner that offers them characteristics that no one substance alone could provide. Metal ions, metal clusters or particles, oxides, sulfides, non-metallic elements, and their derivatives are typically combined in hybrid nanomaterials through specific interactions that enhance their functional properties in a synergistic manner. Electrostatic interactions, dispersion interactions, H-bonding, and other intermolecular interactions may all play a role in the assembly of hybrid materials, from the formation of molecules to nanoscale binding, self-assembly, and microstructuring [27]. The general classification of hybrid materials is shown in Figure 5a. Hybrid materials are now categorized into three types based on their utility, as illustrated in Figure 5a: (i) structurally hybridized materials, (ii) functionally hybridized materials, and (iii) chemically hybridized materials [28,29]. Furthermore, hybrid materials that are formed through weak chemical interactions, such as Coulomb forces, hydrogen bonds, and dipole-dipole forces are classified as Class I hybrid materials. On the other hand, Class II hybrid materials are formed by combining the constituent components through strong chemical linkages, such as covalent or ionic-covalent interconnections.

In the last two decades, a variety of synthesis techniques have been used to rapidly advance the design and preparation of hybrid nanostructured materials with various architectures and dimensionalities. These hybrid materials could be from 0D nanoparticles to 2D nanosheets, including core–shell hybrid nanostructures (for example, MOF-derived NiSe@C nanocomposite [30]), nanoparticle-based hybrid nanostructures (such as Graphene–silver hybrid nanoparticle [31]), well-defined heterostructures (for example, CoZn-Se@N-MX heterostructured hybrid [32]), hierarchical heterostructures (e.g., hierarchical MXene/TMC (SnS, NiS, and MoS_2) [33]), and so on. The techniques for constructing these superior materials, together with nanostructuring, nano-/micro-combination, hybridization, pore-shape control, configuration design, surface modification, and composition optimization, are key challenges for researchers.

Figure 5b provides a summary of the synthetic mechanisms and methods used to create hybrid materials, enabling the creation of custom materials with predetermined features for particular applications [34,35]. Route A is a traditional sol-gel process that employs specific bridged, polyfunctional precursors and a hydrothermal synthesis process to create homogenous molecular organic-inorganic compounds. This approach is highly versatile and can be used to synthesize crystalline microporous hybrid solids such as zeolites and Metal-Organic Frameworks (MOF) [36]. Route B, on the other hand, utilizes self-assembling techniques to create various inorganic nanocomposites or hybrid networks that are templated by organic surfactants. This method provides a high degree of control and enables adjustment over the hybrid interfaces, resulting in a wide variety of nanocomposites. Route C involves assembling or dispersing well-defined nanobuilding blocks (NBB) that retain their integrity in the final product. These NBB can be organically pre- or post-functionalized nanoparticles, metallic oxides, chalcogenides, clays, and layered double hydroxides that can intercalate organic components. Route D combines the use of NBBs and self-assembly methods to obtain hierarchically organized materials in terms of structure and function. This approach enables the tailoring and fine-tuning of the mechanical, optical, electrical, thermal, and chemical properties of hybrid materials, allowing for the development of specialized applications using the synthesized materials [37].

3.1. Applications of Hybrid Nanostructured Materials as Electrodes in Batteries and Supercapacitors

To be an efficient electrode in energy storage devices, materials must adhere to certain standards. For example, the cathode material in a lithium-ion battery (LIB) or a sodium-ion battery (NIB) must exhibit a high free energy of reaction with lithium or sodium to achieve a high voltage, as well as have large interstitial spaces within their crystallographic structure to accommodate a significant amount of Li or sodium ions. Moreover, electrode materials must have a high level of electronic conductivity, be non-toxic, and not react chemically with the electrolyte [38]. When used as an anode, the material should show low voltages (0.0–1.0 V) and usually control the energy density, cycle life, and power density of the cell [39].

Capacitance and charge storage are two criteria that depend on the kind of electrode materials used in supercapacitors. According to the energy density formula, the specific capacitance of the electrode material and the potential difference between the positive and negative electrodes determine the energy density of a capacitor. Developing porous nanoelectrode materials is one of the most effective ways to increase the energy density of supercapacitors. Porous nanoelectrode materials can increase energy density by increasing specific surface area, which increases specific capacitance. This highly efficient method of enhancing the performance of supercapacitors could revolutionize their use in a variety of applications. For example, using carbon materials is more promising due to their high surface area, low cost, availability, and electric conductivity. Surface area, electrical conductivity, electrode wetting, and the permeability of electrolyte solutions are just a few of the variables that have a significant impact on the electrochemical performance of electrode materials [40]. Another approach is to build hybrid/asymmetric supercapacitors, which can improve the overall device performance.

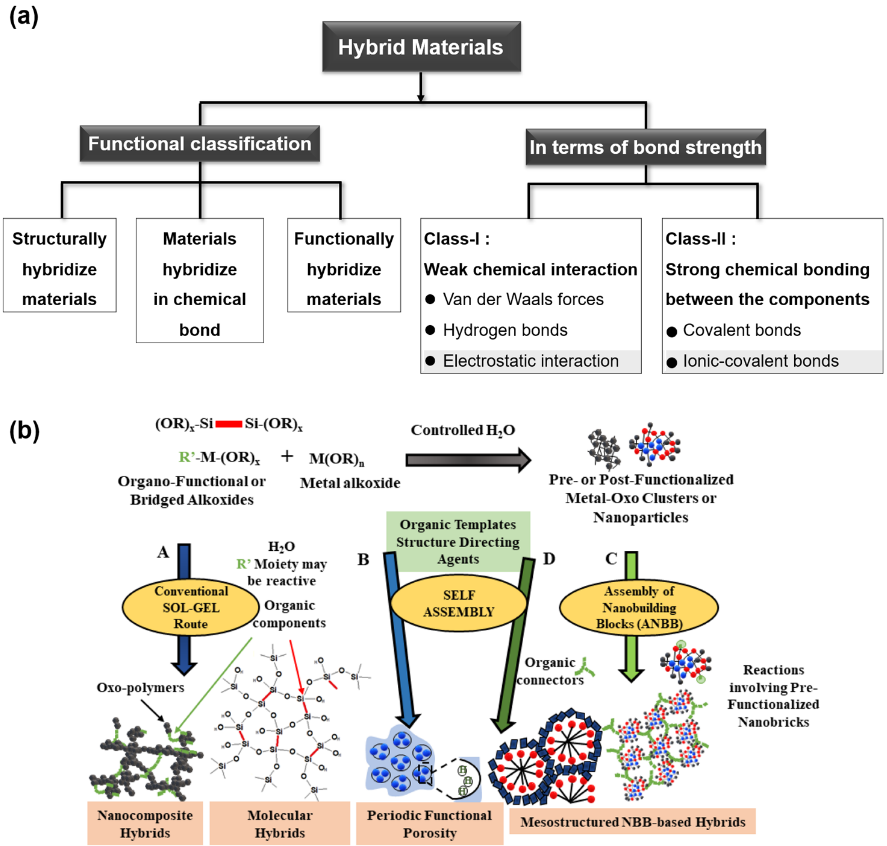

Figure 5. (**a**) The general classification of hybrid materials and (**b**) several general approaches for the design of hybrid nanostructured materials prepared by various routes (A: homogenous molecular organic-inorganic materials, B: self-assembling procedures into account, C: assembling of well-defined nanobuilding blocks and D: combination of self-assembly and nanobuilding block approaches). (**b**) is reprinted/adapted with permission from Ref. [34] 2021, MDPI.

In addition, energy storage materials are critical for the efficient, clean, and adaptable use of energy, as well as the exploitation of renewable energy sources. As a result, they encompass a wide spectrum of materials and have received a lot of attention, in all stages from research to industrialization. In particular, the electrode materials of batteries and SCs with high charge density, cycling strength, rate capability, and stability of batteries and SCs are of utmost importance [41]. The area of the electrode that will be reached by ions/charges may be a particularly crucial factor in determining battery or SC performance [42]. The operational, thermal, and electrical characteristics of devices are largely determined by the electrode material and electrolyte used. The electrodes, for example, must have high conductivity, high temperature stability, long-term chemical stability, high corrosion resistance, large surface area per unit volume and mass, environmental friendliness, and low cost [43,44]. Therefore, to meet the emerging demands, numerous multifunctional hybrid nanostructured materials are being explored at the present time in order to improve the energy density and power of next-generation storage devices. As an example, applications of some of the hybrid materials with promising performances as an electrode in batteries and SCs will be discussed in the following sub-sections.

3.1.1. Carbon-Based Electrodes

Among the diverse electrode materials, carbon compounds have been widely used due to their exciting properties, such as low cost, massive surface area, excessive electric conductivity, availability, non-toxicity, chemical/thermal resistance, environmental friendliness, and excessive stability [45–48]. Nanostructured porous carbon materials have a large surface area, which allows for the addition of functional compounds, such as oxidative groups, hydroxyl groups, or nitrogen, in order to maximize electrode performance. Their surface changes have been demonstrated to allow for increased capacity and electrochemical activity, which is beneficial for the electrolyte. Carbon-based electrode materials also have longer cyclic stability, high performance rate, better safety, and are more cost-effective than pseudo-capacitive electrode materials. When used as electrodes in SC cells, conducting polymers and transition metal oxide-based nanomaterials may result in lower power densities and compromise their long-term stability. To address these limitations, carbon-based asymmetric or hybrid materials are widely used as electrodes in over 80% of commercially available supercapacitor devices [45]. Activated carbon (AC) [49], carbon nanofibers [50], templated carbon [51], carbon aerogels [52], carbon nanotubes (CNTs) [53], graphene [54], and carbon composites are all examples of carbon materials [55] which have been used as electrode materials for batteries and SCs.

CNTs with metal oxides, or conductive polymers based on various sorts of core–shell hybrid nanostructured materials as electrodes, have presented outstanding electrochemical performance, promising constancy, longevity, flexibility, and almost 95% capacitance retention at around 1147.12 mF cm^{-2} at 10 mV s^{-1} (or more), even after 10,000 cycles [56–61]. Their outstanding performance may be due to the formation of interfacial linkages and a conductive grid between the CNTs and substrate molecules within the composite. For example, a nanocomposite of γ-Fe_2O_3/CNT has exhibited great reusability, with calculated capacity of 1186.8 $mAhg^{-1}$ after 400 cycles [56].

As an electrode, graphene has been proven as another potential candidate, due to the existence of a strong van der Waals interaction among its layered structure. Additionally, a highly porous structure with a wide surface area and very good conductivity are its primary characteristic properties, which makes it an ideal material for energy storage applications [62]. Graphene is composed of graphite and has excellent mechanical, electrical, and optical properties. Unfortunately, it also has some practical limitations, including low electron/lithium-ion transport between sheets, which results in poor anode electrochemical performance. Additionally, graphene's large specific surface area can lead to agglomeration between sheets, reducing its effective area and capacity. Due to these constraints, graphene is typically studied as a hybrid or nanocomposite compound, or in a modified form. However, because of its excellent mechanical properties, graphene can also be used as a conductive carrier and to connect active materials, preventing the destruction of electrode structures [63]. Graphene-based three-dimensional (3D) conductive networks may improve electron and ion movement within electrode materials [64]. Several studies have reported the use of reduced graphene oxide (rGO) as a hybrid component with other metal oxides; for example, an NiO/SnO_2/rGO composite has been employed as an anode for Na-ion and Li-ion batteries, which showed a specific capacity as high as 800 mAh g^{-1} at a current density of 1000 mA g^{-1}, even after 400 cycles [65]. Another study has reported that an as-synthesized porous graphene nanoribbon foam displayed greater stability up to 10,000 cycles, maintaining a specific capacity of 123 mA h g^{-1} [66]. Additionally, three-dimensional graphene sponges and aerogels have exhibited a high specific capacitance (~1100 F g^{-1}) at the current density of 10 A g^{-1}.

Activated carbon (AC), as another carbon-based material, has also presented very good performance in the manufacturing of power storage systems, due to its intense conductivity and large surface area [67]. A Li-ion battery exhibited high specific capacity (1351 mA h g^{-1}), with great stability even after 300 cycles, when a sulfur-incorporated AC composite was used as an electrode material [68]. Contact between AC and sulfur decreases the gap among inter-layers, enhancing Li-ion transport as well as energy storage capacity. A mesh-like structure of fullerene (C_{60}) connected by covalent bonds has been shown to be another promising carbon material [69]. The fullerene-coated nanocomposites have exhibited outstanding specific capacity over 2000 mAh g^{-1} with high stability, both as anode or cathode [70,71]. These types of resources may lead to a revolution in determining workable methods for the innovation of cost-effective energy storage devices. The electrochemical properties of various carbon-based hybrid nanostructured materials as electrodes in various energy storage devices, such as batteries and supercapacitors, are listed in Table 1.

Table 1. The electrochemical performances of carbon-based hybrid nanostructured materials as electrodes in various energy storage devices.

Application in Supercapacitors					
Materials	**Capacitance (Fg^{-1})**	**Energy Density ($Wh\ kg^{-1}$)**	**Power Density ($kW\ kg^{-1}$)**	**Retention %/Cycles**	**Ref.**
N, B co-doped -GO	885	23.23	872	80/10,000	[72]
$NiCo_2S_4$/graphene aerogel	704.34	20.9	800.2	80.3/1500	[73]
MoS_2 NS-polypyrrole -rGO	1942	39.1	700	78.6/3000	[74]
SWCNTs/TiO_2	144	20	10,000	95/10,000	[75]
Bio-C/MoS_2	945	157	80	92/10,000	[76]
SnO_2/PCN electrode	799	138	53	95/500	[77]
rGO/MXene-PPy composite	408.2	11.3	500	91.2/10,000	[78]
Ge_4Se_9/RGO/FCNTs	440	32	1071	83/5000	[79]
CoP/CoO@PrGO	402	4.2	785	100/10,000	[80]
Application in Batteries					
Materials	**Discharge Capacity ($mAh\ g^{-1}$)**	**Current Density ($mAh\ g^{-1}$)**	**Retention %/Cycles**	**Application**	**Ref.**
NiS@C	435	50	99.9/500	Mg^{2+}/Li^{+} battery	[81]
F-CuS-CNT hybrid	479	165	85.5/100	Mg^{2+}/Li^{+} battery	[82]
MWCNTs@N-doped-C@CoS_2	1590	100	99.9/250	Li-S battery	[83]
NG/C@Si/CNF hybrid	1346.20	100	97.8/100	Li-ion batteries	[84]
2D Si@SiO_x@MpC	1239	100	99.94/600	Li-ion batteries	[85]
NG/SiO_x/NG hybrids	545	200	99/450	Li-ion batteries	[86]
TC-RGO-CNT hybrid	1401	50	99%/150	Li-ion batteries	[87]
VSe_2@MWCNT hybrid	319.6	50	99.7/200	Na-ion batteries	[88]
C@porousSi/rGO composite	825.7	40	90.4/100	Li-ion batteries	[89]

3.1.2. Metal–Organic Framework (MOF) Electrodes

MOFs (metal-organic frameworks) have shown to be promising materials for efficient energy storage systems [90–92]. MOFs are composed of metal sites and organic linkers. The metal sites are often ions of transition metals, alkaline earth metals, or lanthanides, while the organic linkers are typically multi-dentate molecules with N- or O-donor atoms (e.g., pyridyl, polyamines, carboxylates, and so on). As a result of the presence of highly organized and tunable metal nodes and organic linkers, MOFs offer various unique compositional and structural advantages. Due to their large specific area and low density, MOFs are considered to be very promising electrode materials for new–generation rechargeable batteries and SCs [93–95]. Additionally, various porous MOF nanoarchitectures, such as 0D nanoparticles, 1D nanorods, 2D nanosheets, and 3D hierarchic structures, can be developed through the precise management of MOF growth following re-crystallization, surface management, and size confinement. Moreover, the assembly of MOF crystals into superstructures, or the template-guided growth of MOF-based materials, is expected to allow for increasingly diverse options in this field [94]. In relation to the efficient utilization of MOFs in energy storage devices, the versatile insertion of counter-components, such as polymers, carbons, ionic liquids, and solid inorganic compounds, has inspired the design of new MOF architectures with improved storage capabilities [96].

3.1.3. Halide Perovskite Electrodes

Halide perovskites have recently been deployed for electricity storage in lithium-ion batteries and photo-rechargeable batteries [97–101]. Three-dimensional perovskite structures have been utilized in various fields of energy storage applications, including metal-ion batteries and SCs. Ionic defects within the structure of perovskite materials have a major impact on transport properties such as ionic diffusion and ionic conductivity. These properties are important in the consideration of perovskite materials in energy storage applications [99]. The application of perovskite materials as electrode materials in SCs has been limited, due to their low specific surface area and poor catalytic properties. In recent years, several research groups have reported that transition metal oxides containing a perovskite structure can overcome these limitations. For instance, TMOs with a 3D–perovskite structure—namely, $NiMnO_3$ oxide—have been synthesized via a low hydrothermal method. As an electrode material for SCs, it showed a high specific capacitance of 99.03 F g^{-1} and excellent cycle stability (77%) after 7000 cycles [102].

3.1.4. Transition Metal Oxide and Its Nanosheets as Electrodes

The promising family of transition metal oxides (TMO) and mixed metal oxides has been considered appropriate for use as active electrode materials in batteries and SCs, due to the multiple oxidation states and ions they possess, resulting in superior specific capacitance. Moreover, hybridization with graphene and other materials has gained incredible interest due to their large surface area and excellent electrical conductivity, which is very important for promoting a Faradaic redox reaction [103–105]. In addition, various nanocomposites of TMOs—such as RuO_2, NiO, MnO_2, Mn_3O_4, V_2O_5, Co_3O_4, and their corresponding hybrid materials—have widely been used as electrodes in energy storage studies. For example, the growth of TMO nanoparticles on highly porous carbon nanotubes, graphene, activated charcoal, and carbon fibers has been reported, with a theoretical capacitance of 1300 to over 3500 F g^{-1} [106–111]. Moreover, the incorporation of carbon components can remarkably enhance the overall capacitance. Additionally, increasing conductivity—and, therefore, stability in cycling—makes these composites important for energy storage applications.

Beyond the above, many promising electrode materials have been developed with diverse 2D-nanosheet structures for the design of energy storage devices. Among them, transition metal oxides (TMOs) nanosheets (NSs) [112], transition metal dichalgogenides (TMD) [113], MXene NSs [114,115], layered double hydroxide (LDH) NSs [116,117], and MOFs NSs [118,119] have been shown to present promising characteristics. The hybridization of 2D inorganic NSs with counter nanomaterials/NSs can provide a viable means to investigate high-performance anode materials with extended surface zones, extended voltage windows, increased electrical conductivity, and improved capacitance, in particular. These points of interest of 2D inorganic NSs render them promising candidates in the development of effective cathode materials for different auxiliary batteries and SCs [120,121]. Two–dimensional (2D) inorganic NS-based terminals are regularly synthesized through certain manufacturing methodologies, such as intercalative hybridization, stacking control, core–shell engineering, surface securing, and deformity control [120]. As an example, a 3D $NiCo_2O_4@MnO_2$ nanohybrid has been synthesized utilizing a two-step electro-deposition strategy, which displayed a progressive particular capacitance of 913.6 F g^{-1} with a high capacity of 37.5 Wh kg^{-1} and an extreme control thickness of 7500 W kg^{-1} [122]. Another report has found that a 1 mol% La-doped $Ni(OH)_2$/CNT nanohybrid electrode exhibited a capacitance of 2731 F g^{-1} at 1 A g^{-1} and good capacity retention of 84% at 5000 cycles [123].

3.1.5. Conducting Polymer Electrodes

Conducting polymer- (CP), such as polyaniline (PANI-), polypyrrole (PPy-) and polythiophene (PTh-) based nanocomposites have attracted vital interest due to their physical phenomena, reaction behavior, and promising potential for versatile applications ranging from environmental rectification, energy storage, novel catalysts, and so on. The conductive behavior of polymers is the main reason for their utilization in the development of SCs and

Li-polymer batteries. CPs possess a delocalized π-electron, obtainable redox conditions, and controllable physical assets, which have made them the ultimate choice in the design of various energy storage devices [124]. The sole limitation of CPs is their poor cyclic stability due to architectural demolition of the polymer electrodes, leading to a poor charge/discharge capability [125]. Therefore, the addition of various metal oxides/hydroxides/phosphates and/or CNT graphene to CPs may produce more promising nanostructured electrode materials [126]. For example, according to Shao et al., polyaniline (PANI)/graphene quantum dots/graphene was coated on commercially available compressed nonwoven towels and used as an electrode in SC. The composite material displayed a high specific capacitance of 195 mF cm^{-2} at 0.1 mA cm^{-2} and a high level of stability at 96.5% retention 6000 cycles later [127]. Furthermore, ternary composites such as carbon $LiFePO_4$/PANI have a substantial function as cathodes with increased capacity. The active carbon-based polyaniline composite was chemically oxidized and introduced into the $LiFePO_4$ cathode to improve the low theoretical capacity of 170 mAh g^{-1}. At 10 C, the carbon-$LiFePO_4$/PANI composite electrode outperforms the carbon-$LiFePO_4$ electrode by 26% [128]. Furthermore, the outstanding elasticity of CPs can also facilitate the manufacturing of flexible tools for wearable electric arrangements [129,130]. For instance, Table 2 lists selected conducting polymer-based hybrid materials that are used as electrode materials in batteries, and SCs. Although the progress of varied polymer-based nanocomposite materials has provided considerably more consistent energy storage capacities, beyond this, their fabrication cost and limited performances in some cases until now has been restricted in the consideration of large-scale applications.

Table 2. Applications of conducting polymer-based hybrid materials as an electrode in energy storage applications.

Applications in Supercapacitors				
Electrode Materials	**Capacitance (Fg^{-1})**	**Power Density (kW kg^{-1})**	**Retention %/Cycles**	**Ref.**
MnO_2/PANI/rGO QD	423	640	85/2000	[131]
PANI/S,N:G QDs	2524	2250	100/1000	[132]
PVA-GQD/PEDO	291.86	984.4	98/1000	[133]
TBN–Py CMP/SWCNT	430	-	99/2000	[134]
3D $NiCoO_2$-PPy	1037	465	89/7000	[135]
$LaMnO_3$@CC-Ppy	862	-	66/3000	[136]
Applications in Batteries				
Electrode Materials	**Discharge Capacity (mAh g^{-1})**	**Current Density (mAg^{-1})**	**Retention (%)/ Cycles**	
Polymer/CNT hybrid films	142.3	500	74.6/300	[137]
Poly(Te-BnV) anode	502	-	100/300	[138]
Poly(pyrene-tetraone Sulfide)	697.1	335.4	82/500	[139]
TEMPO-Methacrylate Copolymers	1110	-	99/500	[140]
Metal-organic conjugated polymers	1164	-	99/1500	[141]

4. Hybrid Energy Storage Device (HESD)

As it has been discussed in the above sections, the development of some hybrid nanostructured materials have shown promising results in various energy storage devices. Beyond this, the performance of most of the devices is restricted either by their power or energy capability for energy storage because of above mentioned limitations of the employed electrode materials [142,143]. Therefore, a novel form of hybrid energy storage device (HESD) using the benefits of both battery-type and capacitor-type electrode materials has been reported at first in 1999 by Stepanov et al. [144]. This type of HESD has a high energy density and power density compared to other types of energy storage devices such as traditional batteries and capacitors [145]. As a result, the hybrid energy storage device (HESD) that combines battery-type and capacitor-type electrode materials is one of the most promising next-generation energy storage systems. The basic principle behind the development of this kind of device is some characteristics of batteries and supercapacitors. Especially, batteries provide options with low power density (<1 kW kg^{-1}), high specific energy (30–200 Wh kg^{-1}), shorter lifecycle, lower self-discharge capacity, and lower prices. On the other hand, SCs present less specific energy (<20 Wh kg^{-1}), high specific power (~10 kW kg^{-1}), quick charging, longer time periods (~100,000 times), and high self-discharge capacity. Thus, the combination of batteries and SCs can serve to utilize the complementary properties of each alternative. These HESDs are regarded as one of the most promising energy storage systems for future applications because they inherit the high-power density and long cycle life of supercapacitors and the high energy density of the secondary battery [146]. It is crucial to obtain a perfect match between the positive and negative electrodes since HESD has both battery-type and capacitance-type electrode properties. If the two electrodes are well-matched, the overall performance of HESDs will be enhanced. As shown in Figure 6a, a perfect match between the positive and negative electrodes can result in improved energy density and power density, as well as a longer cycle life.

HESDs can be mainly categorized into two types; (i) asymmetric supercapacitor (ASC), and (ii) battery-supercapacitor (BSC). BSCs are systems in which one electrode stores charge using a battery-type Faradaic process, while the other electrode stores charge using a capacitive mechanism [147]. ASCs are systems with two separate capacitive electrodes. The schematic diagrams for ASC and BSC are displayed in Figure 6b,c, respectively. Based on the electrolytes used, BSCs can be further classified into lithium-ion BSCs (LIBSCs), sodium-ion BSCs (NaIBSCs), acid BSCs (ADBSCs), alkaline BSCs (ALBSCs), and other types. For instance, Li et al. developed a unique LIBSC that used activated carbon as the anode electrode and a composite of $LiMn_2O_4$ and graphene (LMO-MSs@GNSs) as the cathode electrode [148]. According to the electrochemical test results, the LMO-MSs@GNSs composite LIBSC demonstrated an energy density of 38.8 Wh kg^{-1} at a power density of 12.6 W kg^{-1} with an organic electrolyte in the voltage range of 0–2.3 V. After 500 cycles, the capacity was maintained at 90.4% at 2 C. One direction for the development of HESDs is lead-acid batteries. The cycling performance of the electrode material was significantly enhanced by the use of a graphite current collector in H_2SO_4 in a PbO_2 film that was supported by graphite. The completed device can be charged and discharged 3000 times at 10 C, yielding an energy density of 27 Wh kg^{-1} [149].

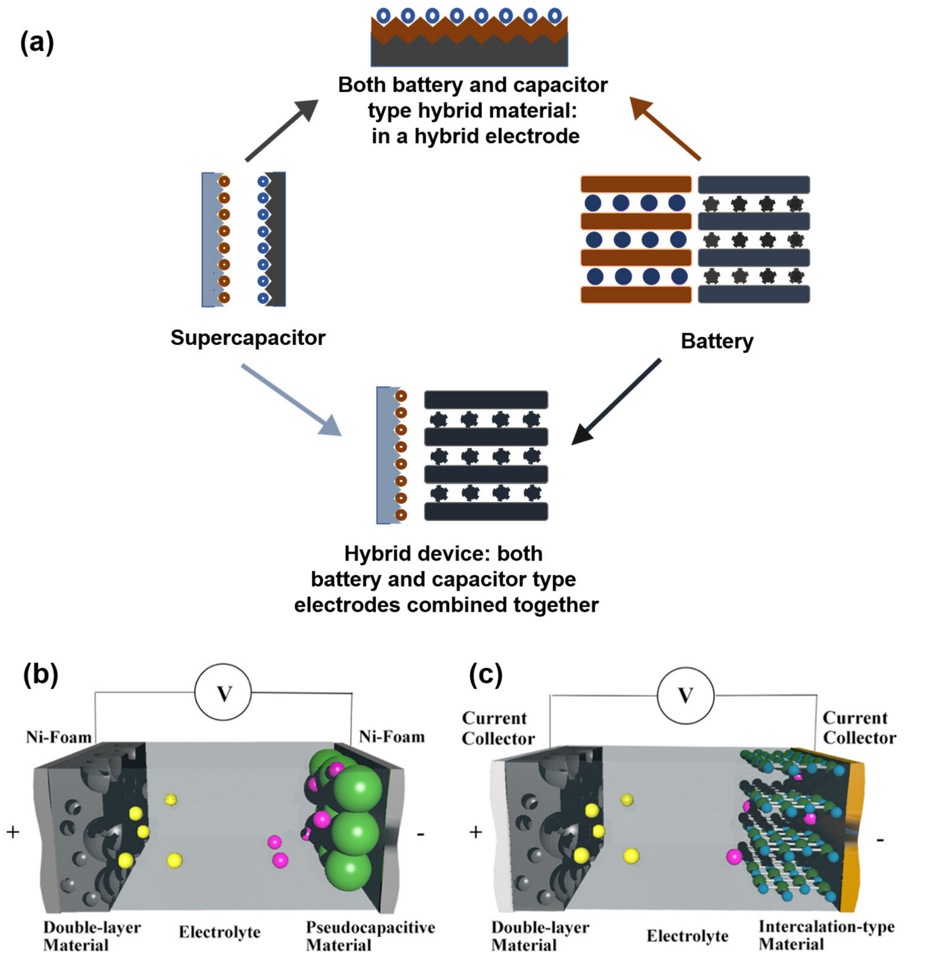

Figure 6. (**a**) Schematic representation of the development of a hybrid energy storage system through the combination of hybrid materials as electrodes for both supercapacitors and batteries. Different types of hybrid energy storage devices (**b**) asymmetric supercapacitors. (**c**) Battery supercapacitors. (**b**,**c**) are reproduced with permission from [150]. Copyright 2019 Elsevier.

According to the various energy storage mechanisms, battery-type materials can be broadly classified as intercalation-type, conversion-type, and alloying-type materials. The electrochemical redox reaction of H^+, OH, Li^+, and Na^+ ions from their respective nanocomposites, such as $LiMn_2O_4$, $LiCoO_2$, $LiFePO_4$, and $LiTi_2(PO_4)_3$, is the energy storage mechanism of the intercalation-type electrode. Activated carbon, carbon nanotubes, and graphene are examples of porous carbon materials with a large specific surface area that are typically used as double-layer electrode materials. Transition metal compounds, e.g., MnO_2, RuO_2, MXene, etc., conductive polymers, and heteroatom-doped carbon materials are frequently used as pseudocapacitance materials. It is crucial to achieve a perfect match between the positive and negative electrodes since the energy storage device combines several charge storage techniques and has properties of both capacitance- and battery-type electrodes. A well-matched HESD can lead to enhanced overall performance. The mass matching method, widely adopted in battery research, is used to match the positive and negative electrodes' capacities at their respective current densities [151]. However, HESD research is still in its early stages, there are certain obstacles as well as opportunities: due to the charging transfer kinetics, the conductivity of pseudocapacitance and battery-type electrode materials is relatively poor, limiting the rate performance. In addition, commonly-used capacitance-type materials such as AC have a very low capacity, making electrode fabrication and matching extremely challenging. Therefore, much work is still needed to develop a HESD that combines high power and energy density.

5. Conclusions and Future Perspective

There is a growing global demand for batteries that are lighter, smaller, more powerful, and efficient, in order to meet the requirements of modern energy storage devices. For instance, lithium-ion batteries are widely used for electronic devices, electric vehicles, and portable electrical equipment. However, many components of battery cells need improvement, leading to numerous options for research and development in the field of high-power lithium-ion cell batteries. The performance of a battery or supercapacitor is significantly influenced by its electrodes and electrolytes. Research has shown that nanostructured hybrid materials are highly promising for fabricating high-performance supercapacitors and batteries, as they can regulate morphologies, surface areas, and electronic properties for energy storage. Therefore, the development of novel electrodes, including those that are micro- and nanostructured, has shown potential to meet these expanding needs. Additionally, several synthesis techniques have been employed to increase their energy storage capacity, including the manufacture of electrodes made from a variety of materials such as conducting polymers, transition metal oxides, and carbon. Furthermore, modifying carbonaceous materials structurally has been used as active electrodes in both batteries and supercapacitors.

Supercapacitors are suitable for applications that require rapid charging and discharging, high cyclic stability, high power delivery, and extended cycle life. Although supercapacitors' energy densities are substantially lower than those of the rechargeable batteries that are now in use, it is anticipated that supercapacitor technology has a long way to go before it can achieve battery-level energy densities to replace it.

Efficient energy storage systems have been the subject of recent studies. However, the goal of developing high-capacity energy storage devices to meet the growing global energy demands is still far from being achieved. (i) The literature suggests that reasonable alterations in the geometry of electrode materials can enhance their electrochemical behaviors. Therefore, a thorough understanding of the electrode-electrolyte interface is essential to design new electrode materials and solid-solid or solid-liquid interfaces. (ii) Nanostructured materials with optimized electrochemical properties are more appropriate for use in battery technologies. Therefore, very effective nanostructured hybrid substances need to be advanced in the future to address imminent power storage issues, mechanical strength, and outstanding conductivity, enabling future power storage functions in batteries and SCs. (iii) The synthesized materials must be inexpensive, highly pure, and easy to produce, and the synthesis process must be adaptable to an industrial setting. (iv) The degradation of the electrode material with use is another crucial factor. To ascertain the electrodes' usable specific capacity and cyclic resistance, a series of chronoamperometric experiment is necessary to prevent mechanical deterioration of the electrode structure resulting from changes in the electrode's volume. (v) To advance further, it may be necessary to use green electrolytes such as ionic liquid electrolytes, which have high electrical conductivity and a large voltage window. (vi) As for increasing the efficiency of electrodes, more and more new hybrid materials must be explored for the resolution of issues related to material structure changes during electrochemical processes, to increase electrochemical efficiency, and facilitate the use of these materials in more demanding energy applications.

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