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Sustainable Materials and Technologies 41 (2024) e01111

Contents lists available at ScienceDirect

Sustainable Materials and Technologies

journal homepage: www.elsevier.com/locate/susmat

Supercapacitors and rechargeable batteries, a tale of two technologies: Past, present and beyond

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ARTICLE INFO

Keywords:
Hybrid
Asymmetric
Supercapacitor
Rechargeable battery
Energy storage

ABSTRACT

Supercapacitors and rechargeable batteries are energy storage devices where the performance strengths of one are traditionally the weaknesses of the other. Batteries benefit from superior energy storage capacity while supercapacitors possess higher power rates and longer cycle life. The rapid adoption of these devices in electric vehicles and grid energy storage applications is driving their further development and production. Accumulating and comprehending the current knowledge of both device technologies shall serve as a foundation for the progress in future research and development within these two distinct fields that share common goals. Therefore, in this review, we aggregate the supercapacitor and battery energy-power performance trends from over the last 18 years, to construct a projection of where the technologies could be heading in the coming decade. We specifically discuss the influence of each of these technologies in the energy storage landscape and their effect on hybridization research. Projections of the trends suggest that by 2040 the best-performing asymmetric and hybrid supercapacitors could be comparable to commercial battery technologies that are currently under development, in terms of energy density (ED). In terms of power density (PD), battery technology can achieve performance comparable to certain electrical double layer (EDL)-based supercapacitors. For some applications, we foresee that the two devices will continue to hybridize to fill the energy-power gap in a way that the penalty to PD for enhanced ED becomes insignificant. This expected improvement may eventually reach a saturation point, suggesting that once a certain level of ED is achieved, any further enhancements of the metric only lead to severe trade-offs with PD, and vice versa. The saturation observed in these technologies has also prompted an exploration of new pathways, with a notable emphasis on sustainability, to achieve high performance using renewable materials and methods.

1. Introduction

In the last few decades, a significant amount of research has been dedicated to energy storage technologies and their materials [1,2]. It is a well-recognized research area that is essential for addressing future concerns about the environmental impact of current batteries and for meeting the rising energy needs in continually advancing power-hungry technologies. The need for more energy led to the development of a modified capacitor called supercapacitors (or ultracapacitors or electrochemical capacitors). Numerous supercapacitor materials and device designs have been investigated and published in this sector during the last few decades, with a notable increase in the number of publications. Much of the literature is dedicated to the improvement of the electrode [3–13] and electrolyte materials [6,8,14–16]. The main targets of material advancements are carbon-based nanomaterials [9,10,17–27], metal oxides, metal-organic frameworks (MOFs) [28–30], and two-dimensional (2D) transition metal carbide-nitride MXenes [31–34]. Additionally, doping carbon is also a common strategy [35,36] to enable pseudocapacitive behavior and also increase ED. Also, in the quest for environmentally friendly energy storage materials, wood-derived lignin

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https://doi.org/10.1016/j.susmat.2024.e01111
Received 22 April 2024; Received in revised form 17 August 2024; Accepted 5 September 2024
Available online 16 September 2024

and cellulose emerge as a promising carbon electrode source for the future. Secondary to material improvements, the investigation, and development of novel device architectures with advantageous power outputs have been frequently reported [7,37–39].

Consistent with the rise in supercapacitor research interest, battery advancement research is also at the forefront of energy storage technology growth. Batteries are broadly classified into primary and secondary types. Primary batteries are non-rechargeable and provide energy based on the reactants used during manufacturing; examples include zinc-carbon batteries (ZCBs) [40], alkaline-manganese batteries (AMBs), and certain irreversible lithium batteries, commonly used in devices like tape recorders and traditional flashlights [41]. Secondary batteries are rechargeable, allowing for the reversal of chemical reactions on applying a reverse potential, making them suitable for applications in portable electronics (e.g., laptops, mobile phones), electric vehicles (EVs), and grid energy storage. Examples include Li-ion batteries (LIBs), lead-acid batteries (LABs), and nickel-cobalt-hydride batteries (NMHBs) [41]. The research in this field of secondary batteries contributes to a wealth of literature corresponding to the improvements in ED in LIBs specifically through novel electrode materials [42–51] and electrolytes [51–56]. Due to the restricted supply of lithium metal and its potential for harm after disposal, research into recycling techniques for these materials has grown [57–60] and attempts to shift to sodium metal-based batteries, which are widely available, have also grown [61–66]. The development of new ecologically friendly batteries [67–69], their disposal techniques [57,70], and other rechargeable batteries [44,71] have also received greater attention as environmental sustainability concerns have grown. Other batteries, such as Li-S batteries [72,73], Zn-ion batteries [74,75], and LIBs with wood-based carbon [76] are also being actively explored as part of this effort to create more sustainable energy storage solutions. Battery cyclability is a critical parameter, focusing on the ability of batteries to maintain high performance over numerous charge-discharge cycles. Therefore much research also focuses on improving battery rechargeability with high cyclability [77,78].

Technically, batteries have had high ED with penalties on PD and cyclability. In contrast, supercapacitors have high PD and superior cyclability with relatively low ED. The distinctions stem from the way batteries rely on chemical processes occurring on the bulk of the electrodes to store and release energy, as opposed to the primarily electrostatic charge storage on the surface of the electrodes in supercapacitors (Fig. 1a). Efforts are directed toward extending the supercapacitor ED and enhancing the battery PD and cyclability, overcoming their respective practical limitations. A Ragone plot (Fig. 2) depicting the device systems' performance in ED versus PD highlights the gap when these technologies progress separately. Nevertheless, each technology has found potential use in specialized applications depending on their existing strengths and drawbacks. Supercapacitors, for instance, have been used in a wide variety of sectors, such as structural sensors [79], phase array radars [8], AC line filters [80–82], wearable electronics

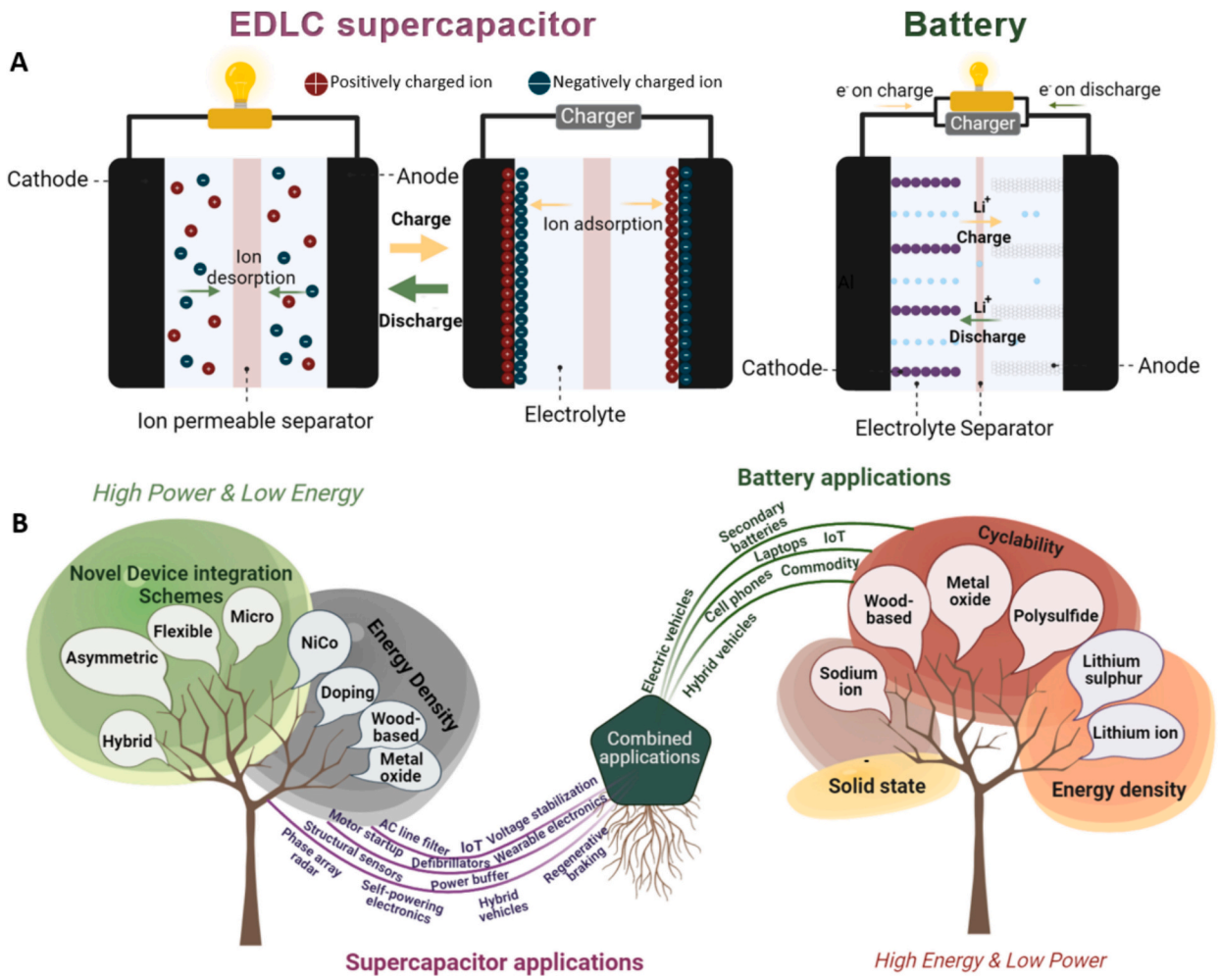

Fig. 1. a) Schematic working principle of supercapacitors and batteries. b) Schematic representing the major focus areas within supercapacitor and battery technologies over the last 18 years targeted at bridging the energy-power gap between the devices. Traditionally limited to specific applications, the merging and evolution of these technologies open up new possibilities for potential applications.

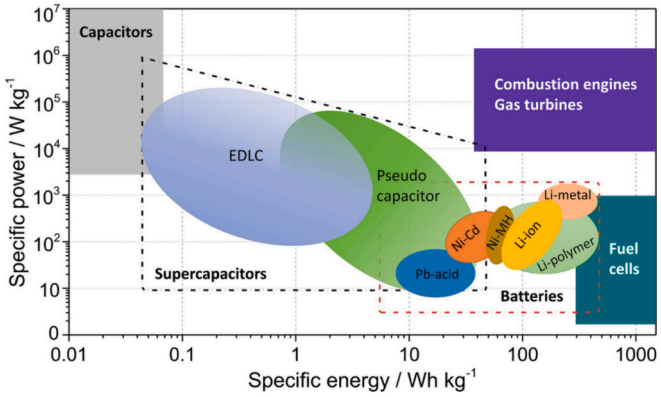

Fig. 2. Ragone plots illustrate the energy storage capabilities of different batteries and supercapacitors (Reproduced with permission, [135]).

[83–87], self-powering systems [88–91], energy-harvesters [92–96], regenerative braking [97–100], IoT [88,101–104] and electric vehicles [105,106]. Their other solid industrial applications include backup power sources, uninterruptible power supplies, wireless alarm systems, and solid-state drives, while also easing grid consumption in commercial buildings during peak demand. Additionally, they play roles in energy storage for renewable sources, automotive start/stop functions, and energy harvesting in rail transportation, highlighting their versatility across multiple industries. Batteries on the other hand are typically found in commodity devices [107] like laptops and cell phones, primary and secondary storage systems [108,109], also IoT [110], electric vehicles [415], and hybrid vehicles [111–114]. To establish an electric-dependent power source for applications such as the automobile sector, it is desirable to have energy storage devices of high ED at higher powers [115,116]. This requirement is a substantial motivation to bridge the energy-power gaps between the technologies [117–122]. The potential for novel applications grows further through the mutual exploitation of both systems (Fig. 1b). In this endeavor to fill the energy-power gap, hybrid device technology emerged [64,122–124]. Supercapattery, supercabattery, supercapacitor-battery hybrid devices, or hybrid energy storage systems (HESS) are some of the terms used in the scientific community to describe the hybridization of supercapacitors and batteries [125–130]. The emphasis of HESS for electric and hybrid vehicles has intensified in the past decade specifically, with extensive research dedicated to configuration, design, energy management systems, and thermal effects [131–134].

Delving into this dynamic landscape, the exploration of HESS technologies holds the potential to unlock a sustainable and energy-efficient future. Although this burgeoning field shows promise in revolutionizing energy storage systems, bridging the gap between battery and supercapacitor technologies presents a formidable challenge. Critical aspects, including configuring optimal hybrid systems, designing advanced electrode materials, implementing efficient control strategies, ensuring safety and reliability, minimizing costs, and mitigating environmental impacts are important. A comprehensive overview of the most cutting-edge materials and devices is crucial for further advancing their technologies to power systems in various fields. This literature review surveys the most active focus areas involved in each of the battery and supercapacitor technologies over the last two decades. Considering both their advantages and drawbacks, an assessment was conducted to elucidate how these technologies function and overlap/interact. This analysis has been conducted to provide the reader with insights into the current state of these prominent energy storage technologies and their future potential development trends. By probing into these focus areas in the fields, the aim is to pave the way for the development of the most highly efficient, cost-effective, and environmentally friendly energy storage materials and systems that can drive HESS's future advancements in electric vehicles, grid storage, and portable electronics.

2. Review methodology

The review statistics in this part describe how the major focus areas (as depicted in Fig. 3) for this assessment were determined through scrutinizing a large set of papers. The most frequently cited works each year over the previous 22 years were used to determine which papers and reviews in both categories were of the highest significance. After carefully reviewing the articles, 62 highly cited experimental and review works were chosen. The most prevalent keywords from these hand-selected publications were gathered and used as the main emphasis areas for this study as a reflection of the state-of-the-art at the time.

To analyze the corresponding field's citation, distribution, and performance trend, the focus area keywords were utilized as search terms in the Web of Science (WoS-https://www.webofscience.com/) search engine. Fig. 3b and d, illustrate the increasing pattern of the total citations in the supercapacitor and battery domains, respectively, for the examined keywords since the year 2000. Li-ion, Li—S, Na-ion, solid-state, metal oxides, polysulfide-based, and wood-based batteries are the major areas of focus in the battery industry. Battery research is increasingly focused on enhancing cyclability and PD close to supercapacitor capabilities. Similar to this, most of the research focused on supercapacitors aimed at increasing the ED [136–142]. Asymmetric [143–149], hybrid [150–155], flexible designs [156–160], metal oxide electrodes [161–166], graphene-based [167–174], nitrogen-doped (N-doped) electrodes [175–180], wood-based electrodes [181–186] and the usage of nickel cobalt electrodes [187–189] were some of their specific research focus areas [190–204]. Asymmetric, hybrid, flexible designs, metal oxide electrodes, N-doped electrodes, wood-based electrodes, and the usage of nickel cobalt electrodes were some of their specific research focus areas [143–204]. Citations in the supercapacitor area indicated that over 75 % of the work is focused on increasing ED, with innovative asymmetric and hybrid supercapacitor device designs accounting for over 47 % of those efforts. The remaining research focused on supercapacitor integration designs, such as flexible/wearable technology and micro-supercapacitors to fulfill different application specifications. As opposed to supercapacitors, batteries are the subject of more varied study, with an emphasis on the environmental friendliness of the device, interest in hazardous chemicals and replacement components in current LIBs, and toxic replaceable Na-ion based batteries. The top citations for metal oxide-based capacitive materials apply also for batteries (Fig. 3d), indicating that efforts to make batteries more resemblant to supercapacitors are exploration-worthy research paths. In the last decade, the development of eco-friendly wood-based carbon electrodes has seen a significant upturn in utilization [203–210,242,243].

In our study, we extrapolate the field's performance trends into the future using a large base of device datapoints from prior studies. The extrapolation of findings from lab-scale prototypes to predict the performance of future commercial products necessitates thorough consideration of factors such as packaging influence and material cost restrictions. It is pertinent to acknowledge that addressing these commercialization concerns falls beyond the purview of this study. However, while analyzing trends, consideration was given to highlighting relevant aspects for readers to navigate effectively.

3. Supercapacitor performance improvement strategies

Up until now, boosting ED has been the primary goal in supercapacitor research. This section examines various strategies attempted so far, comparing their energy-power performance relationships with commercial devices. Standardizing mass reporting is crucial for accurate comparisons, especially with lab-scale electrodes. To ensure precision, light materials like aerogel and graphene are excluded from performance trend analysis due to potentially deceptive data [211]. Nonetheless, their outstanding features, including high cyclability, high-temperature operation, and low self-discharge, are acknowledged in subsequent sections.

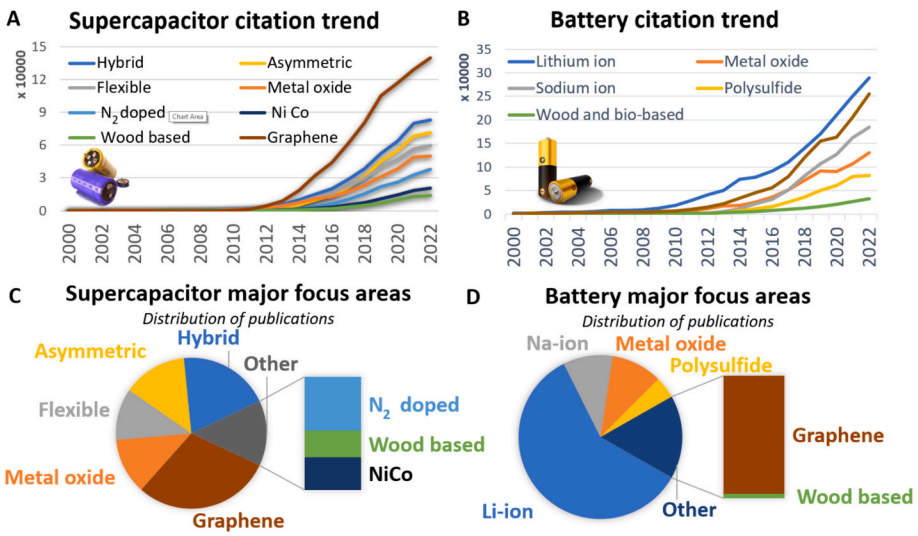

Fig. 3. a) Distribution of publications in the main research areas related to supercapacitors over the last 18 years. b) Citation patterns for the main research focus areas in the field of supercapacitors over the preceding 22 years. c) Distribution of publications in the main research areas related to batteries over the last 18 years. d) Citation patterns for the main research focus areas in the field of batteries over the preceding 22 years.
*(analyzed data from Web of Science).

3.1. Strategies for improving supercapacitor's ED performances

Since the inception of supercapacitors, efforts to improve their energy storage capacity have been consistent. Energy density refers to the amount of energy stored in a given volume or mass of a chosen electrode material for the device. Successful methods for improving ED typically involve modifying electrode materials and creating novel device designs. The quantification of how electrode and device performance rely on energy is comprehended through the given expression for ED as follows.

$$ED = \frac{1}{2}CV_{max}^2 \quad (1)$$

In the equation, C stands for the capacitance and V_{max} is the maximum operating voltage of a device. For energy maximization, increasing the ability to store energy, i.e., an increase in C or V_{max} are obvious ways. The quadratic relationship of V_{max} is more beneficial for boosting energy. Utilizing high-voltage electrolytes and creating innovative device designs are effective strategies identified for increasing V_{max}. For instance, device designs with complementary working potentials from positive and negative electrodes, such as asymmetric and hybrid are constructive to creating high-voltage devices [7].

3.1.1. Achieving optimal performance in symmetric and asymmetric supercapacitors

The standard model of a supercapacitor employs electrical double-layer capacitance (EDLC). EDLC devices of a symmetric type consist of a pair of similar porous carbon material electrodes separated by an electrolyte-soaked separator. They store energy by physical separation of charges at the electrode-electrolyte interface. Tuning the electrodes with easily accessible high surface area contributes to a larger C. However, the V_{max} of these devices is low compared to batteries. Pseudocapacitance, the other type of storage mechanism in supercapacitors, is typically obtained with conductive polymers or metal oxide materials as electrodes. These have higher ED, thanks to their additional C contributions from rapid Faradaic redox reactions and ion intercalation/deintercalation of the electrodes. To manage V_{max}, factors beyond electrode selection must be considered. For example, the V_{max} of symmetric devices with the same electrode in aqueous electrolytes is limited to 1.23 V due to the electrochemical stability window of water-based electrolytes. Utilizing composite electrodes, like MnO_2/carbon, extends the voltage to 2 V even in aqueous electrolytes [212]. The choice of electrolyte significantly influences the working potential. Organic and ionic electrolytes offer options for achieving high V_{max} values, around 2.5–2.7 V and 3.5–4.0 V, respectively [14]. Intentional manipulation of electrode masses of the same type or employing electrodes with different potential windows and charge storage mechanisms strategically extends V_{max} [213,214]. Asymmetric supercapacitors (Fig. 4b), using distinct carbon or pseudocapacitive materials for positive and negative electrodes, achieve V_{max} beyond 2 V due to the complementary nature of dissimilar materials [215]. Choosing matching electrodes is crucial for optimal charge distribution and overall device stability [216,217].

Another classification of asymmetric supercapacitors involves selecting both electrodes with pseudocapacitive materials but with differing redox reactions. For instance, a system utilizing $Na_{0.5}MnO_2$ nanowall arrays as the cathode and carbon-coated Fe_3O_4 nanorod arrays as the anode in an aqueous asymmetric supercapacitor achieves a V_{max} of 2.6 V voltage, showcasing a substantial ED of up to 81 Wh/kg, albeit with a trade-off in PD at 0.65 kW/kg, which is relatively lower for a supercapacitor [218]. PD, defining a supercapacitor's rate of energy charge and discharge, is inherently as high as 10 kW/kg as demonstrated by a supercapacitor with carbon nanosheet electrodes, an EDL material attributed to the rapid ion adsorption and desorption at the electrode surface [219].

3.1.2. Improving electrode and electrolyte synergy in hybrid supercapacitors

Similar to asymmetric supercapacitors, hybrid supercapacitor devices leverage the advantages of two distinct energy storage mechanisms by combining one supercapacitor electrode and one battery-type material electrode. Essentially, hybrids intend to combine the high ED of batteries with the high PD of supercapacitors, aiming at exceptional device performance. In these hybrids, high-voltage battery materials typically serve as the negative electrode for intercalation, while porous carbon functions as the positive electrode (Fig. 4a). Certain electrode materials, including lithium and sodium-based materials, as well as rutile TiO_2, ensure applicability as high-rate negative electrodes due to their impressive rate capability. For the construction of 3 V hybrid supercapacitors, a spinel $LiNi_{0.5}Mn_{1.5}O_4$ LIB cathode with a voltage plateau above 4.5 V demonstrated promising potential. The $LiNi_{0.5}Mn_{1.5}O_4$ /activated carbon hybrid reported an ED of approximately 50 Wh/kg and a PD exceeding 1 kW/kg [220]. Ensuring a high V_{max} without compromising on ED and PD is crucial, necessitating the

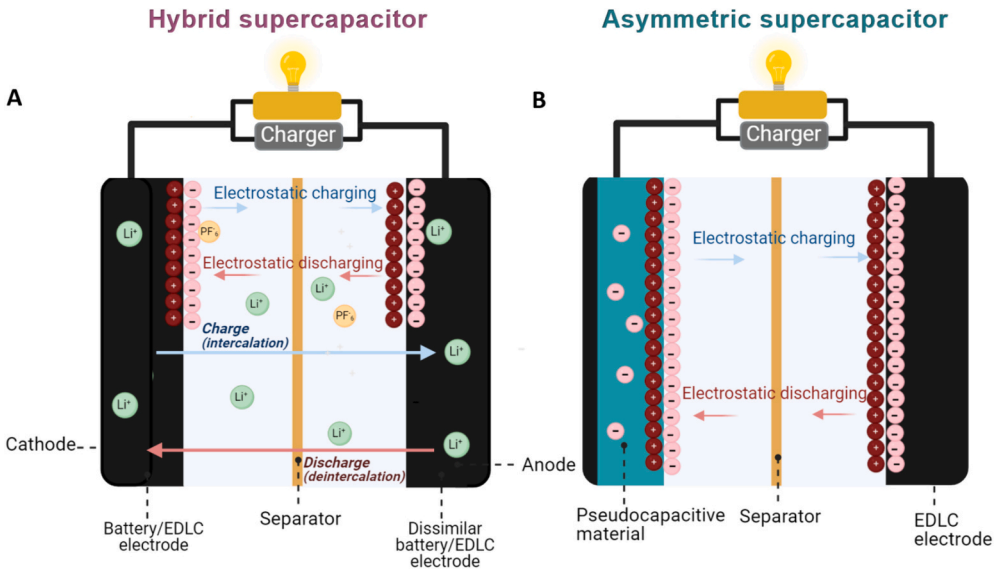

Fig. 4. a) Schematic of the working principle of a hybrid supercapacitor. b) Schematic of the working principle of an asymmetric supercapacitor.

strategic optimization of various components, including the electrode and electrolyte, in energy storage devices. Considering electrodes, nanocomposites represent a breakthrough choice for hybrid capacitors. A hybrid capacitor with active carbon as anode and a porous nanocomposite of La_2NiO_4/NiO achieved a maximum ED of 70.4 Wh/kg even at a V_{max} of 1.8 V [221]. A nanocomposite comprising metal oxide Fe_3O_4 and lightweight 3D graphene as a positive electrode, achieves an impressive ED of 0.15 kWh/kg and a PD of 0.15 kW/kg, accompanied by a V_{max} of 2.7 V [222]. Considering electrolytes, Khomenko et al. demonstrated the development of a high ED Li-ion capacitor (LIC) (Fig. 5a), employing graphite and activated carbon as negative and positive electrode materials, respectively with $LiPF_6$/EC/DMC organic electrolyte. This configuration achieved an impressive V_{max} of 4.5 V, coupled with ultrahigh energy densities reaching 0.1 kWh/kg [223]. Despite ongoing advancements in modifying the structure of porous carbon electrodes, the energy storage capacity of hybrid devices remains limited. To address this, the incorporation of pseudocapacitive materials, such as the newly developed ternary layered carbide and nitride-based 2D materials like Ti_3C_2 MXene, offers a significant boost. For instance, a hybrid device with a positive $Na_2Fe_2(SO_4)_3$ and a negative Ti_2C MXene exhibited an ED of 260 Wh/kg at a PD of 1.4 kW/kg [224]. Further exploration is necessary to uncover the potential of novel 2D materials within the MXene family as electrode materials for hybrid supercapacitors involving lithium and sodium ions.

3.1.3. *Exploring redox-based electrolyte for hybrid supercapacitors*

The strategic selection and optimization of electrolyte materials emerge as another straightforward and effective means to regulate V_{max}. Thus the emerging technology of redox electrolyte-based hybrid devices (Fig. 5b) provides another avenue for boosting supercapacitor ED [227,228]. These devices replace the standard electrochemically inert electrolyte with a redox-active counterpart containing species like transition metal complexes (e.g., VO^{2+}), halide ions (e.g., I^-, Br^-), quinones, phenyl amide, and similar compounds. By integrating Faradaic charge storage into the capacitive mechanism, electron transport occurs on either the positive or negative electrode surface, depending on the redox species. An AC electrode, exhibiting significant redox activity with Na_2MoO_4 species in an aqueous H_2SO_4 electrolyte, achieved a V_{max} of 1.8 V and an impressive ED of 0.38 kWh/kg [229]. While organic and ionic liquid electrolytes are generally suitable for incorporating redox compounds due to their good oxidation resistance and a large electrochemical stability window (2-3.5 V), they are also preferred for their higher chemical stability with lower leakage issues due to lower viscosity [16,230]. Redox electrolytes in general pose challenges of significant self-discharge requiring strategies for mitigation. These include the use of ion-exchange membranes to impede the migration of redox species and the utilization of specific redox species with solid products that can be fixed on the electrode surface during charging processes [231]. Despite the simplicity and effectiveness of using redox electrolytes to create hybrid devices with enhanced energy densities, their intricate energy storage mechanisms pose challenges in understanding the characteristics of each electrode-electrolyte combination.

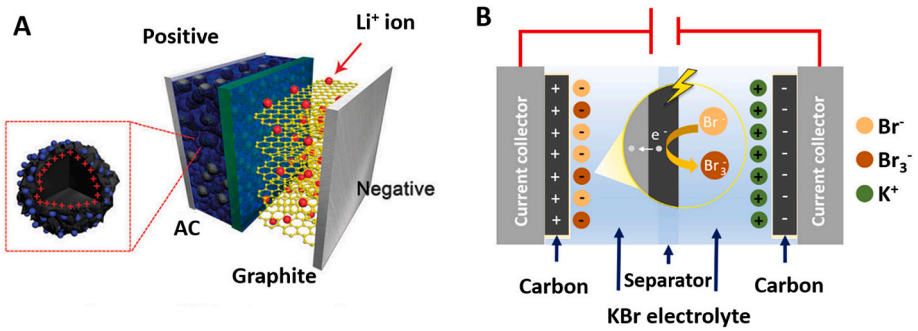

Fig. 5. Schematic of hybrid devices: a) Lithium-ion capacitors. Reproduced with permission from reference [14]. b) Redox electrolyte-based hybrid devices. Reproduced with permission from reference [225,226].

3.1.4. Enhancing performance with N-doped supercapacitors

Introducing redox-active sites on the electrode surface enhances the overall energy storage capacity of supercapacitors, akin to tuning electrolytes. Doping, particularly with nitrogen, is a widely employed method involving the introduction of foreign elements, such as heteroatoms, into the carbon structure. Nitrogen doping enhances electronic properties and electrochemical performance by generating extrinsic defects, introducing additional charge carrier electrons, and improving electrical conductivity [232]. This results in lower internal resistance and faster charge/discharge rates, ultimately enhancing PD. An N-doped carbon nanosheet demonstrated a remarkable PD of 64 kW/kg, retaining 93.2 % of its capacitance after 8000 cycles at 5 A/g [233]. N-doped materials exhibit pseudocapacitance, contributing additional capacitance due to reversible redox reactions involving nitrogen-containing functional groups (Fig. 6). Pyrrolic-N and pyridinic-N are pseudocapacitive, while quaternary N/graphitic-N and N-oxides of pyridinic-N are not. For instance, using N-doped porous carbon nanofibers achieved outstanding energy and power densities of 113 Wh/kg and 105 kW/kg, respectively, at 3 V in the BMIM BF_4/AN electrolyte [234]. Likewise, a symmetric supercapacitor device incorporating an N-doped graphene electrode in a $BMIMBF_4$ electrolyte showed an even higher V_{max} of 3.5 V demonstrating an ED of 55 Wh/kg at a PD 1.8 kW/kg [235]. This performance was attributed to pseudocapacitive nitrogen heteroatoms, particularly pyridinic and pyrrolic species, enhancing electrode wettability. N-doped materials are synthesized through in-situ techniques or carbon post-treatment methods. Post-treatments, including chemical vapor deposition, direct pyrolysis of nitrogen-containing complexes, and hydrothermal carbonization, are more convenient for large-scale production. However, in-situ synthesis offers homogeneous and higher nitrogen doping concentrations.

3.1.5. Designing flexible electrodes and electrolytes for supercapacitors

In the branch of flexible and wearable electronics, the emergence of flexible solid-state supercapacitors stands out as a promising field, primarily utilizing polymer electrolytes to enable flexibility. Notably, asymmetric all-solid-state supercapacitors, employing materials such as free-standing carbon nanotube (CNT)/graphene paper as the negative electrode and graphene/Mn_3O_4 paper as the positive electrode, show an ED of 32.7 Wh/kg [236]. Additionally, composite materials derived from CNT/MOF demonstrate remarkable flexibility alongside a high ED of 59.4 Wh/kg [237]. Another avenue of exploration involves supercapacitors utilizing ion gel electrolytes, where graphene oxide-doped poly(vinylidene fluoride-hexafluoro propylene)-1-ethyl-3-methylimidazolium tetrafluoroborate ion gel as electrolyte achieves an ED of 76 Wh/kg [238]. Furthermore, ionic liquid-based supercapacitors, employing novel ionic liquid gels as solid-state electrolytes, paired with

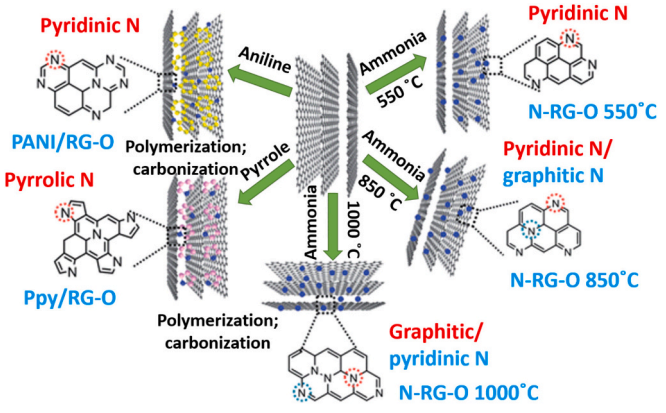

Fig. 6. Schematic representation of N-doped graphene preparation using common ammonia, aniline, and pyrrole precursors by annealing at different temperatures. Reproduced with permission from reference [35]).

MnO_2 and rGO-coated carbon cloth electrodes operating with a V_{max} of 3V, demonstrate a notable ED of 61.2 Wh/kg and a PD of 1.05 kW/kg [239]. These advancements underscore the potential of flexible supercapacitor technologies in enabling next-generation wearable and flexible electronic devices.

3.2. Wood-based electrodes to achieve sustainability goals

Researchers in the dynamic field of energy storage innovations are now exploring environmentally friendly materials to enhance the sustainability of supercapacitor devices. Wood and its derivatives, recognized for their natural abundance, renewability, and exceptional properties, have become focal points of attention. Inspired by the natural microchannels in wood for water transport, researchers have predominantly explored novel wood-based materials for energy storage, aiming to preserve wood's skeleton structure while enhancing conductivity. A noteworthy breakthrough is the development of "wood carbon," achieved through controlled pyrolysis of wood, either with or without activation agents. This process transforms wood into a conducting three-dimensional porous carbon with a high surface area, facilitating efficient electrolyte penetration and providing ample surface sites for energy storage (Fig. 7). These wood-derived electrodes surpass conventional carbon powder electrodes in performance. The latter often incurs high production costs and reduced active material utilization due to additional conductive agents and binders, leading to increased internal resistance. Wood electrodes, highly suitable for the construction of thick electrodes, overcome challenges such as delamination, increased tortuosity, and hindered ion transport faced by carbon powder electrodes with high mass loading [240].

3.2.1. Utilizing carbonized wood for supercapacitor electrodes

A study shows noteworthy electrochemical performance of 800 μm thick carbonized wood electrodes, with significant capacitance contributions [241]. The selection of the right activation procedures, such as CO_2 activation before KOH activation, further enhances the surface area and improves device capacitances [235,236]. Thermal activation of carbonized wood in air, as demonstrated by Zhang et al., at 450 °C for 1 h, effectively opens pores, introduces oxygen functional groups, and transforms the hydrophobic surface into a hydrophilic one, resulting in an activated wood-based carbon electrode thickness of ~1 mm with significantly enhanced current response and an ED of 32.6 Wh/kg at a PD of 17.1 kW/kg [244]. Heteroatom doping with elements like B, N, and P also enhances hydrophilicity, contributing to good electrochemical performances. An asymmetric supercapacitor with N-doped carbon nanosheets from bamboo wood showed 42 Wh/kg (ED) at 4.5 kW/kg (PD). The P-doped wood carbon electrode, with a thickness of 800 μm, incorporates the P element to enhance carbon polarity and redox activity, resulting in a symmetrical supercapacitor device exhibiting an ED of 41.2 Wh/kg at a PD of 26.3 W/kg [245]. Wood's hierarchical porous structure accommodates secondary materials, boosting electrochemical performances in asymmetric models. For instance, an ultrathin MnO_2/C electrode demonstrated a substantial specific capacitance of 480 F/g due to sufficient exposure to electroactive moieties and an expanded layer promoting Faradaic redox reactions. The solid-state supercapacitor derived from this electrode showed excellent performance, achieving a maximum ED of 36.2 Wh/kg at a PD of 18.7 W/kg, facilitated by facile ion access and rapid electron transfer through the conductive pathway [246]. Similarly, Cu nanoparticle-infiltrated wood carbon electrodes act as pseudocapacitive sites, yielding an ED of 123 Wh/kg with a PD of 2 kW/kg [247]. To optimize supercapacitor performance, the focus is on boosting PD while preserving high ED. This is achieved by enhancing electrode electrical conductivity and maintaining stable electron transport during rapid charge-discharge cycles, typically through hybridization with conductive agents. Incorporating a crosslinked PANI fiber network into wood achieved 41 Wh/kg (ED), 68 kW/kg (PD), and a specific capacitance of 800 F/g. Wood's hierarchical

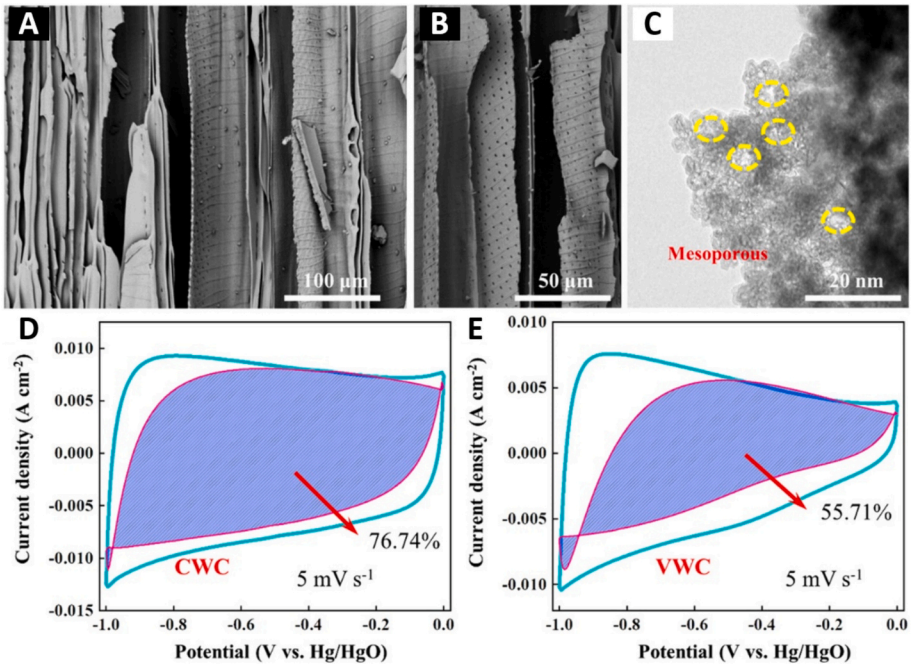

Fig. 7. (a,b) SEM images depicting the vertical-sectional wood-derived carbon (VWC) structure and the presence of pit channels on the surface. Additionally, (c) the HR-TEM image highlights the mesoporous nature (d,e) The capacitive contribution is evident from CV curves obtained at a scan rate of 5 mV/s for both the cross-sectional wood carbon electrode (CWC) and VWC. Adapted with permission from reference [241].

porous structure also prevents conducting polymer shrinkage during increased cycle life [248]. Navigating the diverse architectures and compositions of various wood species poses a challenge in achieving consistent physicochemical properties for wood-based electrodes. Further exploration of wood-based electrodes' performance, considering the discussed major focus areas earlier and complementary electrolytes or device design, is crucial for overall improvement. Possibly, selecting lightweight wood species like balsa and basswood, along with appropriate modification parameters, can yield optimal hard carbon templates for electrode design [249].

3.2.2. *Harnessing lignin and cellulose in supercapacitor electrodes*

Electrode materials derived from wood components - cellulose and lignin for supercapacitor applications exhibit unique strengths and weaknesses. Lignin as the precursor material in porous carbons and hydrochar with outstanding electrochemical properties, shows a superior ED of 46.8 Wh/kg and maintains 22.9 Wh/kg even at a high PD of 25.4 kW/kg [250]. Lignin-based carbon nanofibers (LCNFs), particularly those derived from poplar lignin, exhibit remarkable tensile strength, specific surface area, and capacitance, with an excellent ED of 39.6 Wh/kg at the PD of 5 kW/kg [251]. Lastly, the modified and fractionated lignin-based carbon fibers fabricated by electrospinning stand out with a simple strategy, yielding high-performance energy storage materials with a specific surface area of 2043 m^2/g and a capacitance of 442F/g, resulting in an ED of 56.9 Wh/kg at a PD of 0.39 kW/kg [252]. Yet, a hybrid configuration featuring electrospun activated lignin-derived carbon fiber//Ni-Co-S, while achieving a V_{max} of 1.6 V, demonstrates a modest ED of 30.8 Wh/kg and a PD of 0.8 kW/kg [253]. On the other hand, electrospun N-doped carbon nanofibers (N-CNFs) present a notable asymmetric supercapacitor with high ED (51 Wh/kg) and a maximum PD of 117 kW/kg [254]. Bacterial cellulose -derived N-doped carbon networks, incorporating AC and carbon/MnO_2 hybrid material, demonstrate impressive ED (63 Wh/kg) operating at a V_{max} of 2.0 V, offering an eco-friendly novel device design [255]. The cellulose paper-based asymmetrical supercapacitors with graphite/Ni/Co_2NiO_4-cellulose paper positive electrode and graphite/Ni/AC-CP negative demonstrate superior capacitance, cycling performance, and substantial ED (80 Wh/kg) and PD (25.6 kW/kg) operating in a wide V_{max} of 2.0 V [256]. In another study, they employ a ligand-mediated layer-by-layer assembly to transform cellulose papers into a flexible metallic paper-based supercapacitor with impressive power (129 kW/kg) and high ED (122 Wh/kg) [257]. Utilizing cellulose offers a significant advantage as it is inherently flexible, making it a promising candidate for applications in flexible and wearable electronics. Recently, there has been an increased focus on combining lignin and cellulose fibers due to their complementary individual properties; however, their performances as electrodes in supercapacitors are yet to be explored. Overall, this comparative analysis underscores the diverse potential of cellulose and lignin-derived electrode materials, each offering unique advantages and paving the way for tailored applications in flexible and wearable electronic supercapacitor technology [258].

We have selected a range of devices from each focus area that stand out as top performers in terms of ED, PD, and cyclability, drawing from a data pool of over 400 devices, as detailed in the referenced papers (Table 1). High-performing devices are those capable of enduring numerous charge-discharge cycles without compromising their energy and PD ratings. These chosen devices serve as benchmarks for future advancements in the field. In many instances, incremental performance improvements are compared to older or less efficient devices, ensuring that the benchmark provided is current and reflects the latest developments.

3.3. *Performance trend of major research areas in supercapacitor*

In recent decades, research in supercapacitors has seen significant advancements across various performance metrics, driving innovation in energy storage technologies. This section explores the historical trends and future projections of energy and power density in several types of supercapacitors. Specifically, we delve into the analysis of Ragone plots to compare the performance characteristics of various supercapacitor types and examine how these metrics have evolved. By evaluating these trends, we aim to provide insights into the ongoing developments and future directions in supercapacitor research.

Table 1
Comparison of selected high-performance supercapacitors.

Ref.	Type	Electrodes	Electrolyte	ED (Wh/kg)	PD (kW/kg)	kCycles	Year
[259]	Hybrid	Hierarchical porous carbon-based (Zinc-ion hybrid)	2 M Zn_2SO_4	79	0.4	50	2024
[260]	Hybrid	Ag-WS_2//AC	1 M KOH	50	0.4	1	2023
[261]	Hybrid	HS–NCS@MXene//AC-AHSC	2M KOH	80	1.2	20	2022
[262]	Hybrid	NiCoP/NF-2//AC/NF	2 M KOH	54	2.7	10	2022
[263]	Hybrid	Aqueous sodium-ion Bi_2O_3//MnO_2	1 M Na_2SO_4	71.7 (18.8)	0.4 (3.2)	1.5	2022
[264]	Hybrid	rGO-POM//MXene	1 M H_2SO_4	51	2.8	10	2020
[265]	Hybrid	$NiCoO_2$/NiCoP@Ni nanowire arrays	PVA-KOH gel	40 (24)	0.8(10.2)	2	2020
[266]	Hybrid	MnO_2//reduced graphene oxide	Organic $LiPF_5$	43	30.8	10	2020
[267]	Hybrid	N-doped porous carbon	$LiPF_6$	215(36.6)	65.4 (0.27)	10	2019
[268]	Hybrid	Zn foil//porous carbon	Zn $(CF_3SO_3)_2$	106	31.4	80	2019
[269]	Hybrid	Holey carbon nanolayer//carbon nanolayer	$LiPF_6$	104	65	5	2018
[270]	Hybrid	Mn_3O_4-G // Activated carbon	$LiPF_6$ + EC + DMC	20	6.5	10	2017
[271]	Hybrid	Mesoporous MnO//activated carbon	$LiPF_6$	227	3	5	2016
[272]	Hybrid	MnNCN//activated carbon	$LiPF_6$	103	8.5	5	2016
[273]	Hybrid	NbN//activated carbon	$LiPF_6$	149	45	15	2016
[274]	Hybrid	H-TiO_2 nanowire arrays//activated carbon	$LiPF_6$	94	15	3	2016
[275]	Hybrid	nanosheet carbon//ordered carbon	Organic $NaClO_4$	50	165	10	2015
[276]	Hybrid	NAC//Si-C	Organic $LiPF_6$	141	30	8	2014
[277]	Hybrid	CoO@Ppy//AC	Aqueous NaOH	44	87.5	20	2013
[278]	Asymmetric	$ZnMnO_3$ cubic nanoparticles	PVA/H_2SO_4	182	1.8	2	2024
[279]	Asymmetric	ZnNiCo-LDH/CuO-Cu//N-CNT	2M KOH	117.5	0.58	10	2023
[280]	Asymmetric	ZnCoS NPs@AC//Carbon composite	6 M KOH	43.7	11.2	10	2023
[281]	Asymmetric	NAC//AC	6 M KOH	69	0.69	3	2023
[282]	Asymmetric	Ce-doped MoO_3//N-doped graphene	1 M H_2SO_4	150	0.8	5	2022
[283]	Asymmetric	(Ni—Fe)-P-C@HCNFs//FePC@HCNFs	3M KOH	62.7	8.23	10	2022
[284]	Asymmetric	VS_2-MX-CNT-50 // functionalized MWCNT	0.5M K_2SO_4	60	7.30	5	2022
[285]	Asymmetric	1T-VS_2/MXene// MXene	0.5 M K_2SO_4	41	0.79	5	2021
[286]	Asymmetric	NiCoAl-LDHN-9//AC	6M KOH	36(22)	0.23(4.8)	10	2021
[287]	Asymmetric	CFC-MoS_2//MgAl-LDH-GO	2M KOH	327 (280)	1.5(12)	10	2020
[288]	Asymmetric	NiCo-MOF NSHS//AC	3 M KOH	21	0.75	3	2020
[289]	Asymmetric	$NiCo_2O_4$/C//AC	KOH	375	7.5	3	2018
[290]	Asymmetric	CNF-COOH@MnO_2//AC	$NaHCO_3$	16	7.7	10	2017
[291]	Asymmetric	V-N//V_2O_5	Na_2SO_4	116	25	10	2016
[292]	Asymmetric	Graphene-RuO_2//AC	Na_2SO_4	55	12	4	2015
[293]	Asymmetric	Nanoporous $Ni(OH)_2$ // AC	6M KOH	68	44	10	2014
[294]	Asymmetric	p-BC-MnO_2 //p-BC N-doped	Na_2SO_4	33	284.6	2	2013
[295]	N-doped	Porous carbon	1 M Et_4NBF_4	153	1.24	50	2024
[296]	N-doped (ASC)	Carbon nano onions	2 M KOH	60(25)	1.5(5.5)	5	2022
[297]	N-doped	Carbon nanosheets	6 M KOH	40.5	–	10	2021
[298]	N-doped	Porous carbon	6 M KOH	118	0.2	5	2020
[299]	N-doped	Carbon nanorod	6 M KOH	65	0.35	10	2020
[300]	N-doped	Porous carbon	BMIM BF_4/AN	48	0.75	10	2020
[301]	N-doped	MXene	3 M KOH	22	2.34	10	2020
[302]	N-doped	Porous carbon	KOH	15	1	5	2020
[303]	N-doped	Graphene	KOH	68	0.6	5	2019
[304]	N-doped	Activated carbon	KOH	13	0.5	5	2018
[305]	N-doped	Porous carbon	KOH	25	0.5	20	2017
[306]	N-doped	Carbon nanofibers	H_2SO_4	11	25	10	2017
[307]	N-doped	Activated Carbons	$LiPF_6$	230	1.7	8	2015
[308]	N-doped	Carbon nanosheets	EMIM BF_4	102	–	10	2015
[309]	N-doped	NCNFs	BMIM BF_4/AN [a]	113	105	–	2014
[310]	N-doped	NG hydrogel	KOH	16	205	–	2013
[311]	N-doped	Porous NC nanoplates	BMIM BF_4/AN	133	217	–	2013
[312]	N-doped	Carbon nanofibers	KOH	–	90	3	2012
[313]	N-doped	NG Sheets	BMIM BF_4/AN	55	17	–	2011
[314]	Biomass-based	Lignin AC (corn husk)	1 M TEABF_4/AN	20	0.68	5	2020
[315]	Biomass-based	Lignin AC (jute)	6 M KOH	15	0.40	10	2020
[316]	Wood -based	N-doped porous carbon/Fe_2O_3 nanoparticle	–	30	0.13	10	2023
[317]	Wood -based	N-doped porous carbon/$NiCo_2O_4$ nanosheet	1 M KOH	56	0.35	10	2022
[245]	Wood -based	P-doped carbonized wood	6 M KOH	41.2	0.44	20	2021
[318]	Wood based (asymmetric)	Porous carbon/Mn_3O_4//AC	1 M Na_2SO_4	34.85	0.7	10	2021
[319]	Wood based	Activated carbon	1 M Na_2SO_4	23	0.22	5	2019
[320]	Wood -based	CNTs /Activated wood carbon	1M Na_2SO_4/PVA /H_3PO_4 gel	39.8	0.94	10	2019
[321]	Wood -based	N, S co-doped wood carbon	6 M KOH	15	3.23	5	2018
[205]	Wood -based ASC	Carbonized wood//$Co(OH)_2$@CW	PVA/KOH gel	10.9	0.02	10	2018
[322]	Wood-based	Activated carbons	6 M KOH	9	0.052	–	2015
[253]	Lignin-based BSH	Activated carbon fiber//Ni-Co-S	KOH/PVA gel	30.8	0.8	10	2022
[323]	Lignin-based	Nitrogen containing carbon sphere	1 M SBPBF_4/PC	34	9.4	10	2018
[250]	Lignin-based	Activated carbon	EMIMTFSI	23	25.4	10	2017
[324]	Lignin-based ASC	Porous carbon [a]	1 M H_2SO_4	6.3	1.3	5	2016
[325]	Lignin-based	N-doped porous carbon	PVA-KOH	17	–	5	2016
[326]	Lignin-based	N-doped carbon	EMIM-BF_4	59.8	0.88	20	2016

(*continued on next page*)

Table 1 (*continued*)

Ref.	Type	Electrodes	Electrolyte	ED (Wh/kg)	PD (kW/kg)	kCycles	Year
[327]	Lignin-based	Porous carbon	6 M KOH	~ 24	~ 0.63	1	2016
[328]	Lignin-based	Electrospun lignin carbon fiber/MnO_2	1.0 M $LiPF_6$	84.3	5.72	10	2016
[329]	Cellulose-based	Activated microsphere	4 M KOH	40	15	10	2021
[330]	Cellulose-based	Carbon microtubes	1 M $TEABF_4$/PC	48.3	0.45	5	2018
[254]	Cellulose-based	N-CNF	6 M KOH	51	0.9	5	2015
[331]	Cellulose-based	MnO_2 -CNT-sponge	1 M Na_2SO_4	31	63	100	2011
[332]	Cellulose-based	CNT/conductive paper	1M $LiPF_6$ in EC/DEC	47	200	40	2009
[333]	Symmetric	Porous carbon	Et_4NBF_4	92.6(36.8)	1.24(375)	10	2023
[334]	Symmetric	V_2O_5 nanobelt arrays@RuO_2 nanosheet arrays	1 M $LiNO_3$	174(95.9)	0.45(9)	10	2022
[219]	Symmetric	Hierarchical porous carbon nanosheets	EMIM-BF_4	139 (102)	0.5(9.9)	10	2018
[335]	Symmetric	Porous carbon	EMIM-BF_4	110	4.4	5	2017
[336]	Symmetric	Carbon/MnO_2	H_3PO_4/PVA gel	106	31	5	2012
[337]	Sponge-based	$NiFe_2O_4$/N-CMT-2	6M KOH	50.7 (35.3)	0.25 (5.11)	50	2021
[338]	Composite	MoS_2 NSs @PANI nanoneedle arrays	H_2SO_4	106	106	4	2015
[339]	Graphene-based	N,S co-doped hydrogel	PVA-H_2SO_4	14.8	5.2	7.5	2016
[266]	Hybrid	MnO_2//rGO	Organic $LiPF_5$	43	30.8	10	2020
[222]	Hybrid-graphene	Fe_3O_4/G//3D graphene	$LiPF_6$ with EC/DEC/DMC	147(86)	0.15 (2.6)	0.1	2013
[340]	Asymmetric- graphene	NiCo rGO//rGO	KOH	13	5	1	2019
[341]	Asymmetric-graphene	Graphene- $CuCo_2O_4$//rGO	KOH	45	15	6	2017
[342]	Asymmetric- graphene	β-$Co(OH)_2$//N-doped graphene	solid state	99	18	10	2014
[343]	Asymmetric- graphene	$Ni(OH)_2$-MnO_2 hybrid NS//rGO	KOH	186	0.8	3	2013
[344]	Asymmetric- graphene	Graphene-MnO_2//AC	Na_2SO_4	51	198	1	2011
[345]	Graphene- based	Graphene welded AC	$TEABF_4$/PC	80	0.44	10	2021
[346]	Graphene-based	N-doped GO	3M H_2SO_4	39 (17)	0.38 (7.5)	10	2020
[339]	Graphene-based	N,S co-doped hydrogel	PVA-H_2SO_4	14.8	5.2	7.5	2016
[347]	Graphene- based	Activated-MEGO	BMIM$^+$$BF_4^-$	70	250	10	2011
[348]	Graphene- based	Multilayer graphene films	H_2SO_4	151	35	10	2011
[349]	Commercial	Battery capacitors	–	20-71.4	5	50	2023
[349]	Commercial	Hybrid	–	7-12	4	20	2023
[6]	Commercial	Hard carbon//AC	–	30	10		2016
[6]	Commercial	Graphite//AC	–	15	3		2016
[6]	Commercial	AC//PbO_2	–	30	1		2016

(m1)//(m2) m1 and m2 corresponds to electrodes of same material with different masses.

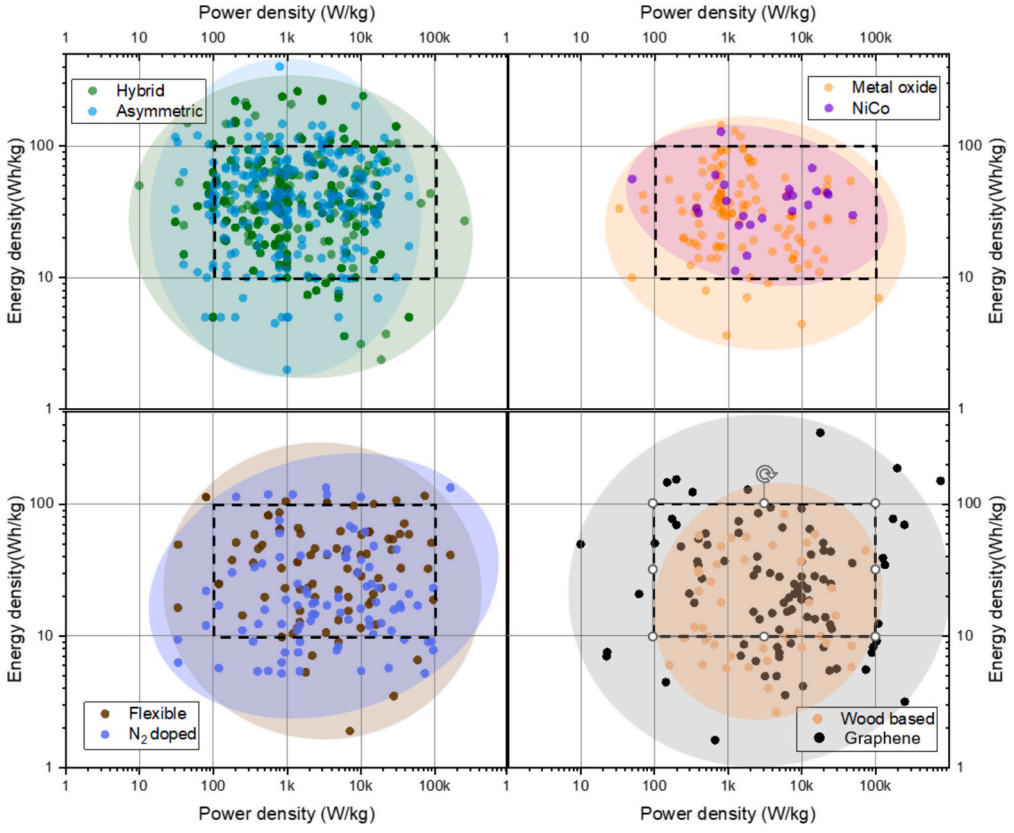

Fig. 8. Ragone plot comparing various lab-scale supercapacitor devices of different technologies in the past two decades.

3.3.1. Ragone plot analysis of various supercapacitor types

This section aims to provide a clear depiction of power-energy relationships in supercapacitor technology by presenting data from over 450 lab-scale devices. Ragone plots, as depicted in Fig. 8, illustrate the concentration of ED with corresponding PD data points within the range of 1-150 Wh/kg and 1-1000k W/kg, respectively. The highlighted dense area termed the 'prime region,' is marked by a dotted black rectangle. Additionally, Fig. 9 presents the performance plot, offering insights into the energy and PD revealing distinctive trends and trade-offs across specific device categories across different focus areas of supercapacitor technology over the years.

These headings are designed to emphasize the use of Ragone plots to analyze and compare the performance characteristics of different types of supercapacitors, focusing on energy density, power density, and overall performance regimes.

In the analysis of supercapacitor performance, as depicted in the Ragone plots, the 'prime region' serves as a reference point. Hybrid and asymmetric supercapacitors exhibit a broader range of higher EDs beyond the prime region, achieving substantial levels at reasonably high PDs. In contrast, devices relying on pure metal oxides and NiCo generally operate within the prime region, highlighting lower EDs but higher PDs compared to their hybrid or asymmetric counterparts. Flexible and N-doped supercapacitors also fall within the prime region, with the latter exhibiting a trend where higher PDs correspond to lower EDs. Graphene-based devices display numerous outliers, while wood-based supercapacitors, though individually unremarkable, align with well-

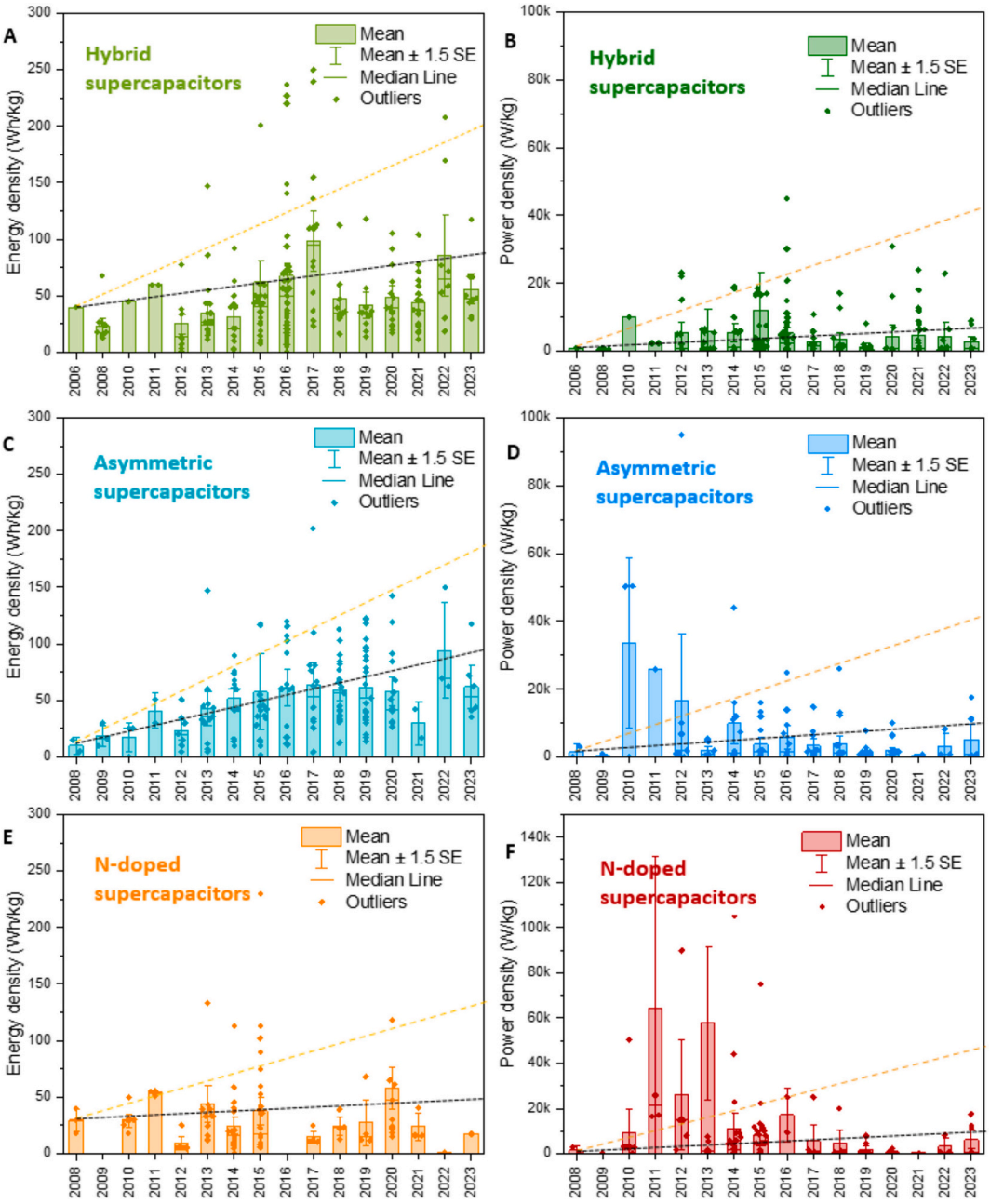

Fig. 9. Energy and PD distribution for hybrid, asymmetric, and N-doped supercapacitor technologies over the years since 2008. The dotted black lines represent a realistic projection of power or energy density for supercapacitor technologies, excluding outliers or overly optimistic estimations based on exceptionally high-performing devices. The dashed yellow lines, on the other hand, represent overly optimistic projections of device performance, considering these outlier devices. The realistic trend was established by aligning with the mean performance metrics of both the ED and PD observed across multiple years. Conversely, the optimistic projection was derived by factoring in the mean performance of outlier devices.

performing devices within the prime region, exhibiting lower EDs for PDs across the spectrum. Despite these analysis trends, all supercapacitor categories face significant power-energy trade-offs, as evident in the projection plot representing varying levels of energy and PDs along the year axis for different focus areas (Fig. 9).

3.3.2. *Evaluating historical trends and future projections of energy and power density of various supercapacitor types*

The trend depicted in Fig. 9 illustrates a general increase in ED over time for most technologies, albeit at the expense of PD as indicated by the PD distribution. Notably, hybrids and asymmetrics consistently demonstrated improvements in both average ED and PD. In contrast, N-doped devices only exhibited moderate ED improvement in recent years while experiencing significant PD losses, possibly due to material performance reaching saturation. Trends were approximated for these technologies to project their future potential performances. Realistic ED and PD projections, excluding outliers, were represented by dotted black lines, while overly optimistic projections considering outliers were represented by dashed yellow lines. Projected ED and PD values were compared with average values for commercial devices in Fig. 10. Rational and inflated estimations were made to compare lab-scale and commercial device performance, accounting for outlier devices. Hybrid and asymmetric supercapacitors consistently surpassed commercial counterparts in ED values while maintaining reasonable PD compromise, indicating potential as a bridge for the power-energy gap. Meticulous supercapacitor design, involving careful selection and matching of electrode materials and optimized properties, is crucial for success in these technologies, balancing energy, and PD for specific applications.

Additionally, energy and PD values reported for lab-scale electrodes often lack standardization concerning packaging, separator, and secondary device components' masses, as well as commercial-level mass loadings. To facilitate an approximate comparison with commercial devices, the histograms in Fig. 10 should be divided by a factor of 3 should be considered [211]. Considering the rational estimations, hybrids and asymmetric technologies show lower PD than even commercial devices, although they demonstrate comparable ED performance.

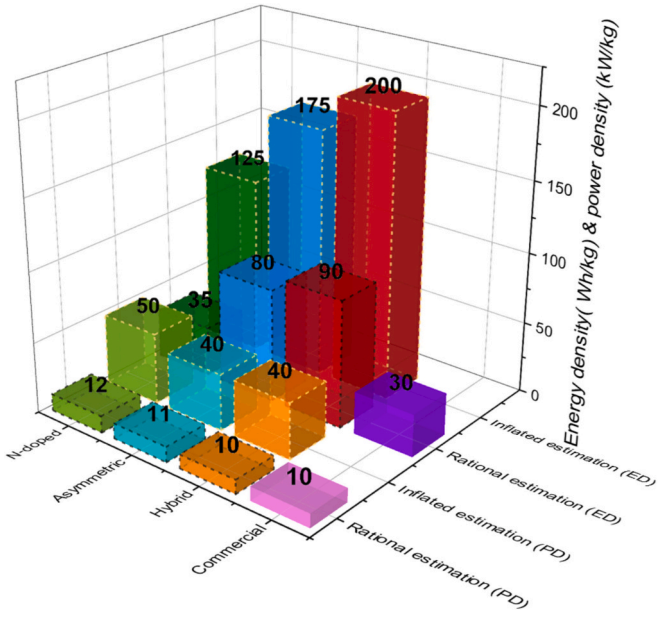

Fig. 10. Rational and inflated energy and PD projections for hybrid, asymmetric, and N-doped supercapacitor technologies compared to current commercial devices. The 'rational estimation' represents a realistic projection of power or energy density for supercapacitor technologies, excluding outliers or overly optimistic estimations based on exceptionally high-performing devices. The 'inflated estimation,' on the other hand, represents overly optimistic projections of device performance, considering these outlier devices.

4. Major research areas in battery and their performance improvement strategies

The most popular and widely used electrical energy storage technology today is batteries. Nevertheless, cyclability, environmental sustainability, recyclability/disposability, and cost are important performance and environmental issues that must be taken into consideration for their continuing growth. As these technologies come together to create hybrid devices, the challenges observed in batteries become more relevant to supercapacitors as well. Consequently, the aim to discuss these challenges will focus on achieving hybridization.

4.1. *Strategies for improving battery's performances*

Since the early 1990s, the Li-ion secondary batteries attained importance due to their high specific or volumetric power, ED, low self-discharge, wide temperature range of operation, and high cycle lifetime, all features which profoundly stand out from other rechargeable batteries [350,351] like lead-acid or nickel-metal hydride. Fossil fuels have a remarkably high ED, and an ambitious target would be to aim at developing rechargeable batteries with equivalent (or higher) ED to cover the demands of applications like electric/hybrid vehicles, enabling a replacement of internal combustion engines with electric motors. The current trend in increased usable ED in state-of-the-art LIBs is too slow to satisfy the practical needs of the surging power demand of portable electronic devices and electric vehicles. The prime reason for the slow advancement in meeting the energy consumption demands is the interrupted energy consumption along with the frequent changes during the battery discharge process. Technological upgrades, optimization of conventional cells' designs, as well as the exploitation of alternative electrode chemistries, have so far represented the most explored strategies for increasing the ED [352,353]. Apart from improvement in-cell technology, the development of new electrode materials [354–357], electrolytes [358–360], separators [361,362], binders [363,364], and current collectors [365,366] is pursued to overcome this problem.

Similarly, understanding battery performance metrics such as C rate and their specific capacity allows researchers and engineers to optimize battery designs and applications for varying charge and discharge requirements, thus maximizing performance and efficiency in diverse settings. In the domain of battery science, the C rate, denoted as *C*, is a fundamental parameter representing the rate of charge or discharge relative to the battery's nominal capacity. It is defined by the ratio of the charging or discharging current I to the battery's nominal capacity $Q_{nominal}$ in Ah:

$$C = \frac{I}{Q_{nominal}} \tag{2}$$

For example, a C rate of 1 indicates a current that can charge or discharge the battery in one hour, while a C rate of 0.5 signifies a two-hour charging or discharging duration.

The specific capacity $C_{specific}$ of a battery refers to the amount of charge it can store per unit mass (m) or volume. It is calculated as:

$$C_{specific} = \frac{Q_{nominal}}{m} \tag{3}$$

The correlation between the C rate and specific capacity underscores the interplay between the rate of charge or discharge and the battery's energy storage capability. Higher specific capacity implies greater energy storage within the same mass or volume, while the C rate influences the effective utilization of this stored energy during charge and discharge cycles. Also, an increase in the voltage (V) of a battery will lead to a corresponding increase in its energy storage capacity, as

described by the provided equation.

$$E_{specific} = Q_{nominal} \cdot V \quad (4)$$

In most literature, batteries commonly utilize capacity (expresseUd in A-h or mA-h) as the primary metric to assess their energy storage capability, while supercapacitors typically rely on energy density as the predominant metric. To establish a correlation between these metrics across these devices, one approach involves multiplying the battery's capacity by its cell voltage to derive the supercapacitor's equivalent energy density. Consequently, the pursuit of materials and systems with higher capacity ($Q_{nominal}$) and a wider operating voltage (V) range is essential for achieving enhanced charge storage capabilities. Both capacity and energy density have been used across various materials and systems in this section, as indicated by the referenced sources. Further, readers may employ eq. 4 to derive $E_{specific}$ for comparison with supercapacitors as both charge and voltage values are deliberately provided.

4.1.1. Advancements in battery cathodes for enhanced ED performance

Lithium is regarded as one of the most promising anode materials in LIBs due to it having the lowest mass-density among metals, ultra-high theoretical capacity, high energy densities, and the most negative reduction potential [367,368]. The major roadblock to the use of Li metal anode in commercial cells is the growth of Li dendrites during the charge/discharge cycles, giving rise to low Coulombic efficiency and potential safety hazards. Various methods such as electrolyte optimization, nano-interface engineering, electrode structure design, separator modification, and the use of solid electrolytes have been developed to mitigate these problems [369]. Still, large-scale production of Li-metal has some serious engineering problems such as mechanical stress by coiling and slitting, electrolyte consumption, and volume swelling stress [369]. Alloying materials such as tin or silicon and metal oxide conversion materials are the optimal candidates among the alternative anodes to achieve both high specific capacities and energy densities [370–372]. Another feasible alternative to conventional graphite anodes is lithium titanium oxide or LTO ($Li_4Ti_5O_{12}$) [373]. LTO is characterized as a highly safe electrode with remarkable thermal stability operating at 1.5 V vs. Li^+/Li, thus eliminating the risk of lithium plating around the anode during charging. This LTO anodes material cycles at an amazingly fast rate with a specific capacity of 170 $mAhg^{-1}$. Hard carbon is emerging as an alternative to graphite-based anode due to its overall superior ability to store Li^+ ions as well as superior fast charging abilities and cycle life performance [374]. In addition, hard carbons have low cost, high thermal stability, they are environment friendly and precursors for making hard carbon anodes are naturally available in abundance [374]. The main challenges in commercializing hard carbon is their structural degradation at higher current and low initial Coulombic efficiency [374]. Disordered materials offer promising features as electrode materials for LIBs due to their compositional flexibility, disordered open structure, structural stability, and redox reaction efficiency [375]. These features enable disordered materials to have superior Li^+ transport and storage properties, volume change-buffering advantage, and high structural reversibility [375]. Significant research and development have been done in the use of graphene and its nanocomposites as an anode material for LIBs. Graphene as an anode exhibits some really promising features, such as minimizing the area of the active material before the nanoparticles are redistributed and generating greater transition rates of ions/electrons [376]. Graphene functions as an anode in two ways: either as a composite material to improve conductivity and reduce volume changes of the other active anode materials or as a supporting material to increase efficiency. Consequently, materials containing graphene and its derivatives (GO, rGO) have superior storage capacity and good rate-performance [376]. There are some serious major issues which are limiting the use of graphene and its composites as an anode material in LIB, for example its coulombic efficiency, cycle life, safety, energy and power density [376].

The prime foci for cathode material research are dedicated to achieving higher specific capacity and/or working voltage for increasing the battery ED while lowering the cost and toxicity. Layered compounds, namely $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ (NMC) and $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ (NCA) have an average operating voltage of about 3.7 V vs. Li^+/Li, higher reversible capacity (about 180-185 mAh/g), and an added advantage of higher thermal stability in their delithiated form with lower Co content than the conventional cathode material $LiCoO_2$ [377–379]. Ni-rich cathodes like NMC811 are emerging as promising next-generation cathode materials due to their higher specific capacity, higher operating voltage, higher energy density, superior thermal stability, and lower cost due to less Co content [380]. However, Ni-rich cathode materials suffer from loss of compositional elements, irreversible distortion of the lattice structure, and structural reconfiguration. These shortcomings of Ni-rich cathode materials can be mitigated by lattice, compositional, and microstructure engineering [381]. Also, Li-rich layered compounds $(1\text{-}x)Li_2MnO_3.xLiMO_2$ (M = Co, Mn, Ni, and Li/M ratio > 1) have garnered attention due to their remarkable capacity which exceeds 200 mAh/g, and simultaneous low cobalt content. Despite that, these materials suffer from structural instability, voltage hysteresis, and decay after prolonged cycling, therefore the current focus is on optimization to overcome these problems [377,382]. Another low-cost and environmentally friendly cathode material is $LiFePO_4$ olivine which is already used in commercial LIBs [383]. $LiFePO_4$ olivine has remarkable chemical and electrochemical stability with respect to other discussed layered-structure materials [384]. Recently, a large number of polyanionic olivines namely $LiFe_{0.5}Mn_{0.5}PO_4$ and $LiCoPO_4$ have been investigated as viable cathodes for next-generation LIBs [385]. These materials have a higher working voltage than $LiFePO_4$ and also a higher theoretical ED than $LiFePO_4$ [386]. Although $LiCoPO_4$ exchanges Li^+ ions at a remarkably high potential of 4.8 V vs. Li^+/Li, their high expected cost, and environmental concerns due to rich cobalt content are major drawbacks. Recently some new classes of polyanionic cathode materials like $LiFeSO_4F$, Li_2FeSiO_4F, and Li_2FeSiO_4 have been reported to show great rate capability, superior capacity, and amazing cycling life performance [387]. Another interesting class is spinel-structured electrodes, namely $LiMn_2O_4$ and $LiNi_{0.5}Mn_{1.5}O_4$, which are Co-free cathode materials suitable for high-energy LIB batteries working at about 4.1 V and 4.7 V vs. Li^+/Li, respectively [388,389]. The current focus of LIB is to achieve the double-high of high energy and power density. This can be achieved by developing cathode materials with high-voltage, and fast-charging capabilities [390]. Researchers are trying to achieve this goal through surface, bulk, compositional and electrolyte engineering.

As discussed earlier, Li metal as an anode material has immense potential for high ED battery systems. The true potential of Li metal anodes is exploited in the Li-air (Li—O_2) and Li—S battery systems [57,73,391–394]. The theoretical ED of Li—O_2 batteries approaches that of gasoline. The major problem with Li—S batteries is that they suffer heavily from the lithium polysulfide (LiPS) shuttle effect. Li—O_2 batteries on the other hand suffer from oxygen/moisture erosion of the Li metal anode. Zinc-air batteries have a remarkably high ED of 220 Wh/kg [395,396]. The zinc-air cell is mainly used as a primary battery. However, recharging is possible by physically replacing the zinc electrodes which is quite inconvenient for practical applications. Important to point out is that many of these modern technologies have a low energy content per volume unit, and are thus not suited for automotive applications, however, for stationary applications the high ED is incredibly attractive. Flow batteries where the chemical energy is provided by two chemical components dissolved in liquids contained within the system and separated by a membrane are in the developmental stage [397,398]. The energy inflow cells are stored not as the electrode material as we observe in conventional batteries but as the electrolyte in row cells and the ED in row cells is a function of electrolyte volume. Commercially Sodium-ion batteries (SIBs) have come up as an alternative for LIBs due to their comparable energy density with LFP chemistry, better safety fast charging/discharging capabilities, and lower cost [399]. One of the

major cell manufacturers, CATL has already launched its first-generation sodium-ion battery in 2021. Other cell manufacturers like Faradion, Northvolt, and TIAMAT are soon going to launch their commercial SIBs. Research has shown that Calcium ion batteries (CABs) can achieve doubled or even tripled energy density as compared to the state-of-art LIBs [400]. Moreover, the cost of CABs will be significantly less than LIBs. The major hurdle in CAB technology is efficient Ca metal anode/electrolyte combinations operating at room temperature and cathode materials for CABs [400].

So far high-capacity cathode and anode materials for high ED batteries have been discussed. However, to tap the full potential of these materials, new electrolytes with high lithium-ion (Li^+) conductivity, as well as high chemical, mechanical, and electrochemical stability, are to be developed. The traditional liquid electrolytes have risks of leakage and combustion of their organic constituents. Another issue that poses safety concerns for future high-energy batteries with Li metal anodes is the growth of lithium dendrite in liquid electrolytes. On this concern, gel electrolytes and solid-state electrolytes are preferred over traditional organic liquid electrolytes due to their reduced flammability and improved stability along with safety [401–403]. Nanohybrid electrolytes are another promising route toward safer, high-performance electrode design [404,405]. In nanohybrid electrolytes, organic (polymers, ionic liquids) and/or inorganic (Li^+-conductive ceramics and glasses) materials are combined to produce a wide range of new electrolytes with interesting chemical, mechanical, and electrochemical properties compatible with different cell chemistries.

For batteries to match the PD of supercapacitors an appropriate electrode material with excellent electrical conductivity, a high surface area, and tailored pore size is envisioned. MOFs are such types of appropriate electrode material that can bridge the gap between SCs and rechargeable batteries in terms of energy and PD [406,407]. They have a controllable morphology with many desirable properties such as low density and copious pores resulting in a remarkably high surface area. Also, MOFs can be tailored by selecting definite metal sites and modifying their pore sizes to balance their contribution to power and energy densities [408–410]. The recent research on MOFs and MOFs-derived carbon materials are promising and they are emerging as prospective electrode materials for energy storage devices having both high energy and PD [29,411,412], because of their well-regulated morphology with a flexible structure, significantly high specific surface area, large pore volume, and accessible metal sites.

In September 2020, a notable advancement in Li-ion battery technology was introduced with the unveiling of new cylindrical cells, denoted as 4680 cells [413]. These cells, depicted in Fig. 11, offer a larger size compared to the previous 2170 cells, enabling a considerable increase in energy storage capacity. 4680 cells have the potential to store five times more energy, significantly enhancing the ED of Li-ion cells. Moreover, a novel tabless cell design, patented concurrently, is anticipated to further elevate the ED of LIBs.

In December 2020, a US-based startup announced a breakthrough in solid-state Li-metal battery technology, presenting its first-ever implementation [414]. This innovation, as displayed in Fig. 12b, incorporates a flexible ceramic material that serves both as a solid electrolyte and an efficient separator. Notably, the performance outcomes released demonstrate nearly double the ED of leading commercial lithium-ion cells. This achievement is attributed to the development of an anode-free design (Fig. 12a), where Li^+ ions migrate from the cathode, forming a flat metal layer (in green) on the opposite side of the separator to establish a temporary Li anode during charging. Conversely, during discharge, Li^+ ions return, ensuring that all lithium contributes to energy storage, thereby significantly boosting the ED of the cell.

Additionally, ongoing research efforts are focused on enhancing anode-less Li-ion cells by modifying negative electrode current collectors to further optimize performance [416].

4.1.2. *Advancements in battery anodes for enhanced PD performance*

LIBs are well known for their high energy densities, but their power densities are less impressive. The LTO batteries are the ones with the highest PD [417]. If conventional gasoline engines are to be replaced, LIBs should be able to deliver higher power densities in addition to their high energy densities. This requires materials with fast lithium diffusivity and rapid insertion and extraction of lithium ions. Various electrode materials have been developed and their structures are tailored so that the LIB performance can be stretched toward meeting the high-power demand of automotive applications. Some of the major efforts to increase the PD of the LIB start with the anode materials. Carbon-based materials have been favored as anode materials owing to their excellent cycling ability and long cycle life [418]. Graphite compounds exhibit a theoretical capacity of 372 mAh/g [419] along with exceptionally high diffusivity of lithium-ions [420] which makes rapid charging and discharging possible, enabling improvement of the available power significantly. Functionalized multiwalled CNTs synthesized using a layer-by-layer technique exhibit a PD of 100 kW/kg and are stable over a thousand cycles [421]. However, the major drawback of such carbon nanotube anode materials is their low reversible capacity of 200 mAh/g. Recently graphene has emerged as a potential anode material for Li-ion batteries. The possible adsorption on both sides of graphene results in a high reversible capacity of 784 mAh/g [422].

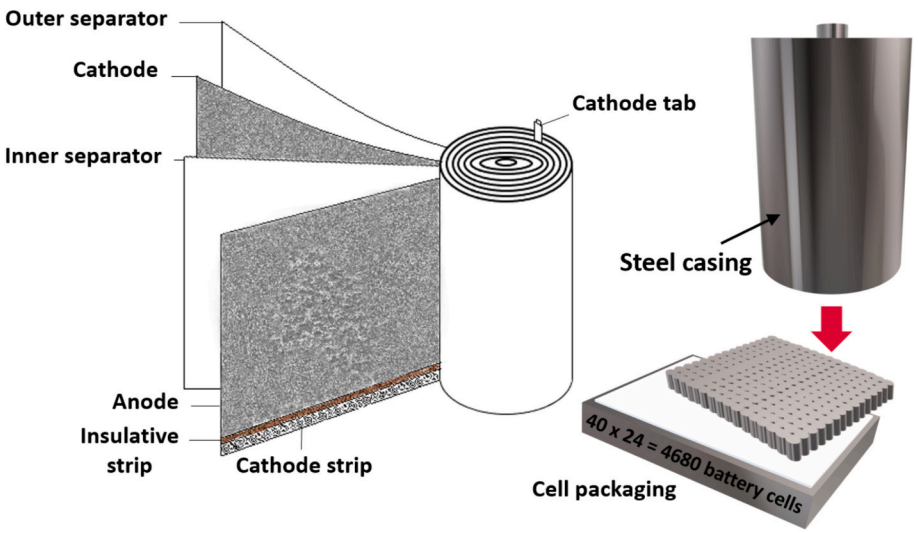

Fig. 11. Tabless cell design [413] with 4680 cylindrical cells.

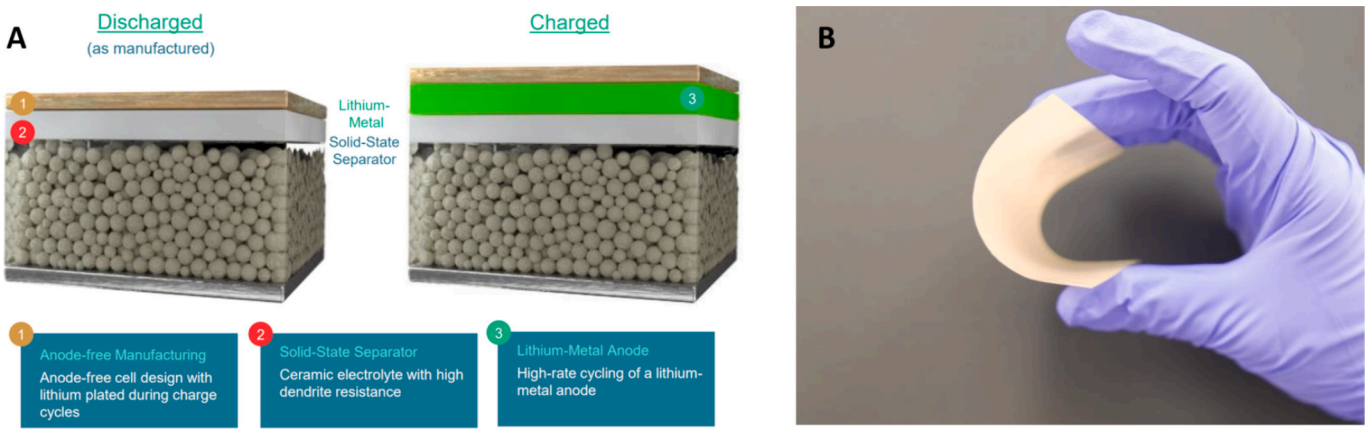

Fig. 12. a) Anode-free cell design [414] b) with flexible separator and solid-state electrolyte [414].

Graphene also has inherently high conductivity which significantly improves the charge transfer. Unfortunately at rates of 3C and above graphene sheets have been reported to block fast Li^+ transport thereby limiting its application to high-energy LIBs [423]. Sputter-deposited amorphous carbon film is another promising anode material having high obtainable capacity, low irreversible capacity loss, and high coulombic efficiency [424]. However, cycling at a high C-rate due to partial diffusion of lithium ions in the film the obtainable capacity reduces. Therefore, to achieve both high power densities and high capacities, alternative anode materials need to be investigated. Pure aluminum, tin, germanium, and silicon or their composites are among the potential anode materials for high energy and improved PD LIBs. Although aluminum-lithium alloy anodes have a high capacity they suffer from rapid capacity fading during subsequent cycling and very slow Li^+ diffusion. Tin is superior to both graphite and aluminum as anode material due to its high theoretical charge capacity of 994 mAh/g. It also has a higher operating voltage and the diffusivity of lithium ions in tin is also very high: 5.9×10^{-7} cm^2/s [425]. The most severe limitation of tin is its poor cycle life due to volume expansion up to 300 % during the alloying and dealloying process [426]. Researchers are attempting to address this problem. A three dimensional porous Sn—Cu alloy has been developed and it exhibits high cycle rates and stable capacities [427,428]. Tin oxides also have a significantly high theoretical capacity of 1.5 Ah/g, high coulombic efficiency, and good cyclability [429,430]. The large surface area of nanofiber tin oxides makes them a better candidate than tin oxide thin films for use in Li-ion batteries. Tin oxide nanofibers can deliver steady capacities over 0.5 Ah/g for 1400 charge/discharge cycles at an extremely high rate of 58C [431]. The reason behind this superior performance of tin oxide nanofibers is their ability to withstand volume changes. Similarly, composites made of SnO_2 encapsulated in carbon nanotubes exhibit improvement in the power densities of Li-ion batteries. The tubular structure withstands the large volume changes during lithium's insertion and extraction [432,433]. Silicon and germanium are also seen as prospective anode materials owing to their superior theoretical capacity of 4.2 Ah/g and 1.6 Ah/g, respectively. Unfortunately, both suffer from three major disadvantages. First, germanium and silicon, both being semiconductors, have poor electrical conductivity which leads to inefficient charge transfer during charge/discharge cycles. Secondly, the diffusivity of lithium ions is significantly slower in them which limits their charge/discharge rate [434,435]. Lastly, both silicon and germanium undergo a massive volume expansion of 300 to 400 % [436–439]. Extensive research is carried out to find ways to overcome these limitations either by doping or nano structuring or by making various nanocomposites [371–373,440].

Next, the cathodes are considered for improving the PD. Cathodes are primarily categorized into three types: lithium metal oxides, spinels, and lithium metal phosphate. The performance of cathode materials is largely governed by structural stability, conductivity, and Li^+ diffusivity. Among the cathode materials, lithium metal oxide $LiCoO_2$ is widely used with a theoretical capacity of 273 mAh/g but due to a limited cut-off voltage of 4.2 V, only 50 % of the theoretical capacity is obtainable. Operating above the cut-off voltage of 4.2 V leads to structural change and dissolution of cobalt resulting in rapid capacity degradation [443]. Moreover, the relatively low diffusion coefficient of lithium-ion in $LiCoO_2$ limits its application for high power performance. $LiCoO_2$ has been doped with lanthanum to enhance its performance, which indeed resulted in an improvement in the stability of the layered structure, enhanced electron transfer-ability, and greater capacity retention [444,445]. Proper nanoscale engineering has effectively reduced the diffusion distance for lithium-ions resulting in superior diffusion kinetics along with high rate capability and high electrode density [446,447]. The performance of cathode material has been significantly enhanced by coating $LiCoO_2$ with CuO nanoparticles [448]. The toxicity and cost of $LiCoO_2$ can be reduced by partial substitution of cobalt with nickel as it also improves structural stability by suppressing the lattice expansion [449]. Mixed nickel and cobalt oxide cathode materials are capable of high-power performance, but they have a short cycle life. While delivering high power a dramatic increase in cathode impedance has been observed [450]. Another promising cathode material that can reduce both cost and toxicity is $LiMn_2O_4$ spinel [451]. Initially, $LiMn_2O_4$ spinel suffered from some serious limitations like structural changes, manganese dissolution, lattice distortions, and microstrain [452–454]. Cationic substitution of Ni in the spinel structure reduced the microstrain and improved performance. The dissolution of manganese has been controlled by coating the spinel electrodes [455]. Coating of spinel cathodes also induces structural integrity thereby improving the cycle life [456]. Nanostructuring of spinels because of better accommodation to the changes in volume and shorter diffusion distance improved their rate capability and power densities, respectively [457]. The final cathode material, i.e., $LiFePO_4$ has low cost, low toxicity, and long cycle life compared to cobalt oxides and other spinel-based materials. $LiFePO_4$ has only one drawback, it suffers from poor conductivity that could limit its application in high-power batteries. However, using smart nanostructuring (Fig. 13), doping, and carbon coating (Fig. 14) this limitation of $LiFePO_4$ has been successfully overcome, making $LiFePO_4$ suitable for high power LIBs [458–462]. The current and future advances in nano-engineered (nanostructured and nanocomposite) anode and cathode materials could result in LIBs capable of delivering both high energy and power densities [463].

4.1.3. Wood-based electrodes to achieve sustainability goals

For more environmentally friendly choices, researchers are exploring the use of hierarchical wood structures as battery electrode material. A

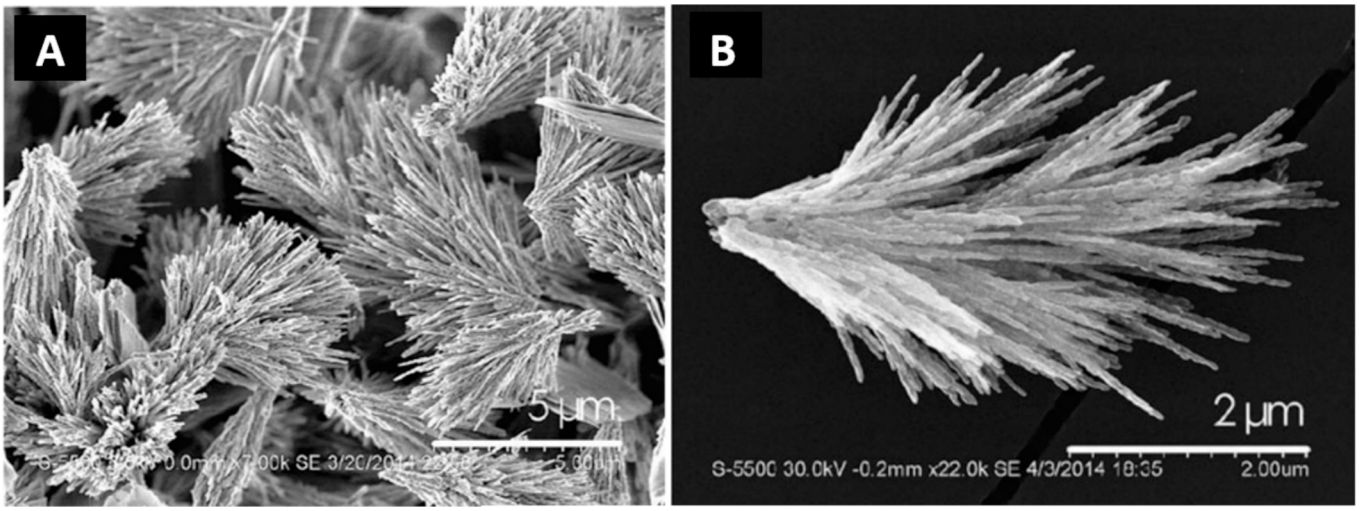

Fig. 13. a) A representative FESEM image showing the uniform $LiFePO_4$ nanoarchitectures composed of self-assembled nanowire-like building blocks. b) A magnified FESEM image of an individual nanoarchitecture. Reproduced with permission from reference [441].

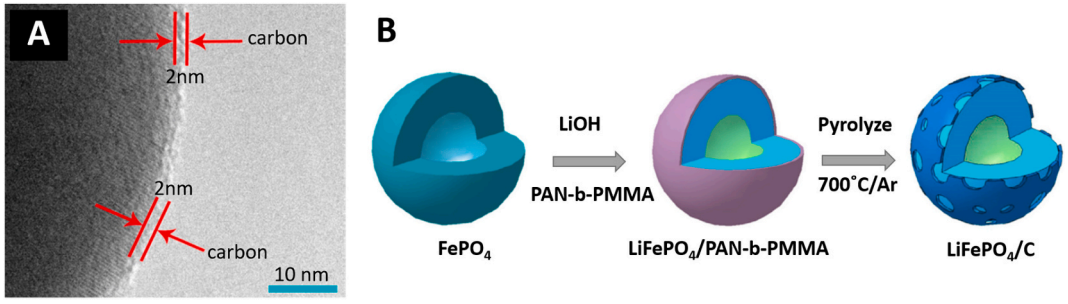

Fig. 14. a) TEM image of $LiFePO_4$/C nanocomposite 10 % PAM-b-PMMA. b) A scheme of the formation process of $LiFePO_4$/C Reproduced with permission from reference [442].

study introduced a composite device made of natural wood fiber, CNTs, and a Sn film, achieving a capacity of 339 mAh/g [464]. Additionally, an electrode consisting of interpenetrating networks of lignin and PPy, derived from lignin extracted from brown liquor, exhibited a capacity of 70 –75 mAh/g [465]. Hard carbon derived from lignin exhibits an impressive reversible capacity of 584 mAh/g, achieved through an increase of advantageous carbonyl groups and large interlayer spacing. They demonstrate significant promise for LIBs owing to their remarkable

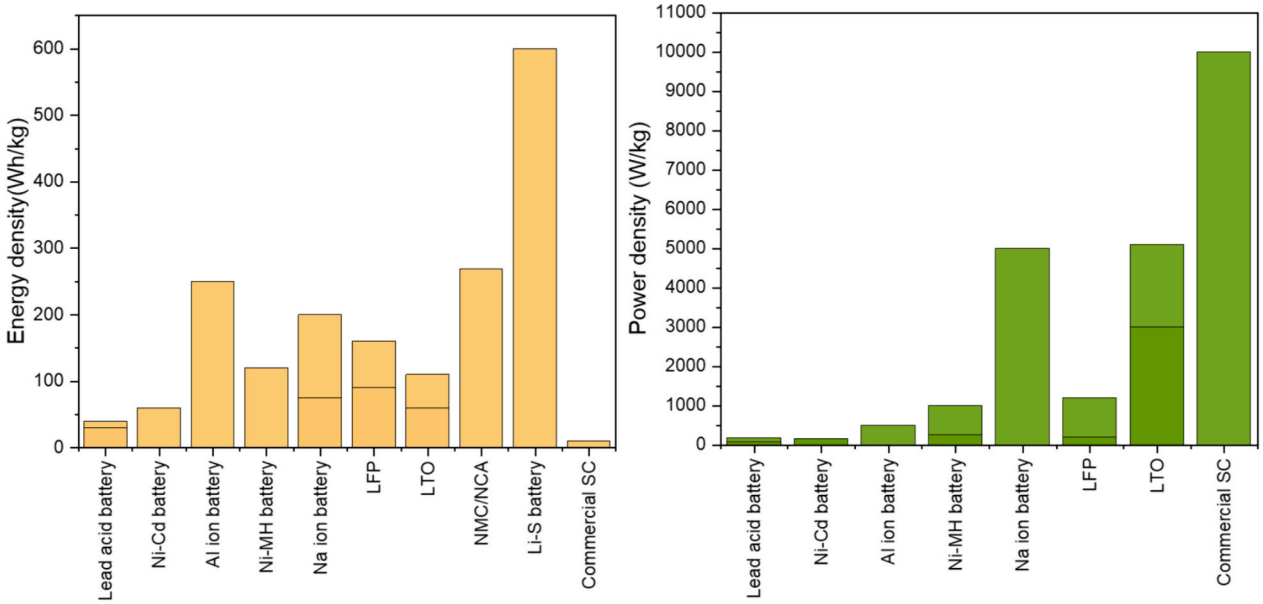

Fig. 15. Energy and PD distribution for different battery technologies.

ability to store Li-ions. In an aim to advance capacity, N-doped honeycomb lignin nanoporous carbon was created via KOH activation and urea doping. As an electrode in Li—S batteries, this material effectively addressed the issue of polysulfide dissolution through its proficient microporous structure. Notably, the composite exhibited a remarkable initial capacity of 1.3 Ah/g at 0.1C and sustained capacity retention with 0.65 Ah/g after 600 cycles [466]. Anyhow, the EDs offered by wood-derived energy devices typically are far lower compared to traditional metal-based systems. A reason is they lack inherent conductivity and therefore need to be integrated with conductive materials, which also presents challenges. Furthermore, research is needed to address issues such as shelf-life stability and self-discharge in biomass-derived energy devices.

4.2. Performance trend of major research areas in batteries

Fig. 15 displays the performance comparison of various battery technologies with commercial SCs as of 2022 [467,468]. SCs give better PDs than any battery technology, as is clear and well-known from the discussions. However, because LIBs have such high EDs, their application has thus far thrived in industries like automobiles. By improving the electrochemistry and mechanical design of the cell at the industrial level, LIBs have been developed further. However, as the supply of lithium metal is limited, there is a need for more readily accessible, inexpensive, and non-toxic materials that result in high power. This search has led to the attention of Na-ion and Al-ion-based batteries. Na-ion batteries perform on average better than modern commercial batteries in terms of energy and PDs. Al-ion batteries operate similarly, delivering high energy at the expense of poor power delivery. However, Li—S batteries, like other LIBs, have environmental problems and are far from being commercially beneficial despite their ability to solve this power output shortfall. No other battery technology is currently competitive with current commercial SCs in terms of power performance. Additionally, it is anticipated that the commercial SC would perform much better with the development of related lab-scale technologies like hybrids and asymmetric SC.

It would have been interesting to include PD performance data for wood-derived electrodes, similar to other battery technologies. However, due to the limited data available in the literature, it was not feasible to include them in the analysis.

5. Systematic comparison of supercapacitors and batteries

Understanding the differences and similarities between supercapacitors and batteries is crucial for optimizing their applications, improving economic feasibility, reducing environmental impact, and driving technological innovation. This comparative analysis will highlight key performance metrics, material compositions, costs, and suitability for various uses, providing valuable insights for selecting the appropriate energy storage technology for specific needs.

Table 2 presents a comprehensive comparison of the operational and functional factors that distinguish the performance and capacity of supercapacitors, broadly classified based on design configurations, from those of standard LIBs in contemporary applications.

5.1. Lifetime and cyclability

Supercapacitors and batteries differ significantly in terms of lifetime and cyclability, each presenting unique advantages and disadvantages. Supercapacitors have a notably longer lifespan than batteries, often lasting 10-20 years under normal conditions, and maintaining 80 % capacity even after 8-10 years of use [472]. In automotive applications, supercapacitors can endure 10-15 years [473]. Their cycle life is exceptional, reaching up to 1 million cycles, and they can be charged and discharged virtually unlimited times with minimal wear and tear.

In contrast, LIBs typically have a shorter lifespan, lasting 5-10 years in vehicles [474]. Their cyclability is also more limited, with a defined cycle life usually around 500 cycles or higher [475]. LIBs degrade more significantly with repeated cycling, and their lifespan is affected by factors such as temperature, charge rates, and state of charge (SOC).

Table 2
Comparison of various supercapacitor configurations with LIB.

	Symmetric SC	Asymmetric SC	Hybrid SC [469]	Lithium battery[a] [470,471]
Charge storage	Double-layer capacitance (electrostatic)	Double layer capacitance (electrostatic) & pseudocapacitance (faradaic charge transfer)	Double layer capacitance (electrostatic) & faradaic capacity (electrochemical)	Intercalation
Electrodes	Carbon materials	Carbon & metal oxides	Carbon & battery materials	$LiCoO_2$ cathode and graphite anode
Electrolyte	Organic (footnote)	Aqueous	Organic	Lithium salt in an organic solvent
Operating voltage	Up to 3 V	Up to 2.3 V	Up to 3.8 V	3.7V
Energy density	3–5Wh/kg	30Wh/kg	100Wh/kg	120-190
Power density	9000 W/kg	5000 W/kg	4000 W/kg	1000-3000 W/kg
Thermal runaway risk	No danger	No danger	No danger	Yes
Operating temperature	-40 °C to +65 °C	-25 °C to +60 °C	-40 °C to +65 °C	0-45 °C
Self-discharge	High (weeks)	Moderate (days)	Long (months)	Less (years)
Operation lifetime at RT	10-20 years	~10 years	5-10 years	2-3 years
High temperature operation [b]	2000 h	2000 h	1500 h	Deteriorates device life
High voltage operation [b]	1500 h	1000 h	1500	Becomes unstable
Cycle life	1000,000 cycles	100,000 cycles	10,000 cycles	>500 cycles
Shelf life	4 years	2 years	3 years	7-15 years
Availability	Commercially available	In research phase	In research phase	Commercially available
Cost/kWh	$100-500	$100-$3000	$2500-3500	$100-170
Current applications	Memory backup	Power applications(check)	EV	UPS, portable electronics, and so on.
Toxic level	Low	Moderate	Moderate	High

[a] The lithium-ion battery family is represented here by the user-dominant $LiCoO_2$ cathode-based LIBs which are generally common for powering portable consumer electronics.
[b] Typically beyond prescribed operating temperatures or voltages.

There is a broad agreement that temperatures above 30 °C accelerate aging [476–478]. High C-rates, which refer to faster charging and discharging, also contribute to quicker degradation [479–481]. Moreover, higher SOC levels usually lead to increased degradation, although avoiding high SOC values can substantially improve the lifespan, as seen in LMO/NMC-based cathode cells [476,478,482,483] [484]. Si-doped anodes, on the other hand, appear sensitive to low SOC values.

While LIBs offer high PD, their potential is consistently constrained by lifespan concerns. Issues such as structural failures from electrode and electrolyte interactions, and the corrosive effects in specific types like Li—S batteries, further limit their cycling performance [394]. Aging models for LIBs range from empirical to semi-empirical and circuit-based, with some attempts at physical-based modeling [476,478,482,483]. Second-life usage of batteries is gaining attention for economic and environmental benefits, but significant performance issues remain [485–487], [488].

Overall, supercapacitors excel in longevity and cyclability, making them suitable for applications requiring frequent charge-discharge cycles and long-term reliability, whereas batteries, despite their higher energy density, face challenges related to lifespan and cycling performance.

5.2. Safety considerations

Supercapacitors are generally considered safer than lithium-ion batteries for several reasons. Firstly, they have a lower risk of thermal runaway because they do not involve flammable electrolytes, making them less prone to catastrophic failures. Additionally, supercapacitors can function effectively across a broader temperature range, from approximately -40 °C to +70 °C (-40 °F to 158 °F), which exceeds the operating range of most batteries [489]. They also generate less heat due to their lower internal resistance, resulting in less energy wasted as heat during charging and discharging cycles. Unlike batteries, supercapacitors store energy electrostatically without chemical reactions, reducing the risk of chemical-related safety issues. Environmentally, supercapacitors are more friendly as they do not contain lead or other harmful substances found in some batteries.

However, supercapacitors still require proper handling and safety precautions. They can operate at high voltages, necessitating careful handling to avoid contact with the main terminals. Their ability to discharge rapidly can pose risks if not properly managed. For safe transportation, supercapacitors should be shipped in a discharged state with a ground strap across the terminals. Moreover, some supercapacitor-based products incorporate isolation circuits to enhance safety when the system is not in use or powered down.

Ensuring the safety of electrolytes is a critical concern in energy storage systems, particularly in applications such as electric vehicles. Solvent-free ionic liquids with low vapor pressure and high thermal stability offer a promising solution to address this concern without compromising energy and power [490]. To fully leverage the electrochemical stability of ionic liquids, specialized electrode materials, and cell configurations become essential [490]. While acetonitrile is a commonly used solvent for supercapacitor electrolytes due to its low viscosity and high conductivity with salts, its high volatility and flammability pose challenges. To mitigate these risks, aprotic room temperature ionic liquids like TMPA–TFSI are introduced, enhancing the safety of the supercapacitor device [491].

In contrast, LIBs present more significant safety challenges. Batteries using lithium metal as anode material are more dangerous compared to those containing acids, mercury, nickel, cadmium, and lead. Water presence in inadequately dried cells or ingress upon loss of mechanical integrity can cause ignition and generate highly flammable hydrogen gas [492]. Dendritic growth of lithium on the Li metal anode can pierce through the separator and reach the cathode, causing a short circuit and increasing the risk of thermal runaway leading to explosive reactions [493,494]. Polyolefin-based porous membranes used as commercial separators are thermally and structurally unstable, shrinking at elevated temperatures and rupturing upon force application. While some commercial separators have been improved with thermally stable coatings, new types of separator materials and structures are being developed to enhance cell safety [495,496] [497–499].

Additionally, volatile, and flammable liquid electrolytes increase the risks associated with LIBs [500,501]. To address these safety issues, lithium metal anodes have been replaced with graphite or carbon-based materials [502,503], and alloys or nanocomposites [504] of lithium metal anodes have been developed to utilize the full capacity of a lithium anode Surface modifiers and better separators are being designed to make Li metal batteries safer. Replacing volatile liquid electrolytes with solid-state [505,506] or polymeric electrolytes [507,508] has significantly improved the safety of LIBs. Thermal runaway is a major safety concern for LIBS. Thermal runaway mitigation and prevention has three aspects: intrinsic safety, active safety, and passive safety. Intrinsic safety is related to the thermal stability of electrode materials, separators, and electrolytes, which has been discussed earlier in this section. Active safety is related to the thermal management of the cell or pack. Coupling Phase Changing Materials (PCMs) with other thermal management techniques particularly to increase temperature uniformity is a crucial aspect of achieving battery thermal management [509]. To stop the uncontrollably rapid spread of heat between cells assembled in a pack, researchers are focusing on developing low thermal conductivity, high thermal stability, flame retardant, and heat-insulating materials. The final aspect i.e., passive safety is related to the early detection of thermal runaway. To achieve the early warning of thermal runaway of the battery, it is imperative to concentrate on the use of gas detection sensors since the electrochemical process occurring inside the battery will produce a huge number of gaseous substances. For implementing early monitoring and early warning technologies for thermal runaway of the battery, the embedded optical fiber sensor or electrochemical impedance meter may detect the internal temperature and impedance of the battery in real-time [509]. External stress factors such as charging at sub-freezing temperatures, vibration, mechanical stress, elevated temperatures, overcharging, and complete discharging are also hazardous for batteries [510,511]. Safeguarding batteries from these stresses has been implemented using electronic protection circuitry in the battery pack and a good battery management system (BMS) [512,513].

Solid-state batteries offer immense promise, particularly in terms of safety. Unlike traditional LIBs that use flammable liquid electrolytes, solid-state batteries utilize non-flammable solid electrolytes, significantly reducing the risk of fire and explosion [514]. This inherent safety feature makes solid-state batteries particularly appealing for applications where safety is paramount, such as electric vehicles and portable electronics. However, while solid-state batteries offer promising safety features, the current body of research and understanding in this field necessitates further exploration and development to achieve successful commercialization [515].

In summary, while supercapacitors and solid-state batteries present enhanced safety profiles compared to traditional lithium-ion batteries, each technology requires careful consideration of its respective risks and handling procedures. The choice between supercapacitors and batteries often depends on the specific application and its operational environment.

5.3. Economic evaluation

Supercapacitors and batteries exhibit distinct cost dynamics in techno-economic analysis, reflecting their initial costs, operational lifespans, and manufacturing trends. Initially, supercapacitors are significantly more expensive per kWh compared to batteries, ranging from $17,000 to $1500 USD/kWh, as noted in [349]. However, their extended operational lifespan of up to 1000,000 cycles, compared to around 1000 cycles for batteries, contributes to their cost-effectiveness

in long-range and heavy-duty applications [349]. Manufacturing advancements, particularly in scalable methods like screen printing and inkjet printing for paper-based supercapacitors, further enhance their potential cost-effectiveness [516]. SC manufacturers have achieved substantial cost reductions of 65 % to 75 % over the past 15 years, with projections indicating an additional 30 % decrease by 2030 [517].

Conversely, battery costs are influenced by materials and manufacturing processes. Lithium iron phosphate (LFP) cathodes are preferred for their relative affordability [518], whereas materials like nickel manganese cobalt (NMC) offer higher energy density but are costlier due to cobalt's expense [137]. Efforts focus on reducing costs through nickel-rich electrodes and exploring alternative materials such as sodium [519–521] and silicon [522]. Manufacturing efficiencies are also crucial; studies predict a decline in battery cell costs from $106/kWh to $64/kWh in the future [523,524]. Moreover, second-life applications for LIBs in electric vehicles are explored to extend their economic value, mitigating costs associated with degradation and interaction with the power grid [525,526].

These factors underscore the complex economic considerations between supercapacitors and batteries, emphasizing the trade-offs in initial costs versus long-term operational savings and the evolving landscape of materials and manufacturing efficiencies.

5.4. Techno-economic analysis

Also, techno-economic analysis of supercapacitors and batteries exhibit distinct cost dynamics. Initially, supercapacitors are generally more expensive than batteries for equivalent energy storage capacity, with costs potentially exceeding those of batteries by up to tenfold. However, over their lifespan, supercapacitors can prove more cost-effective due to their longer operational life and higher cycle count. For instance, research detailed in [527] highlights that while battery-only systems cost 8.4 ¢/kWh, supercapacitor-only systems can cost as little as 2.8 ¢/kWh, and HESS further reduce costs to 2.6 ¢/kWh, illustrating their potential economic advantage in certain applications. Aging costs are a significant consideration, with supercapacitors primarily affected by calendar aging, contrasting with batteries where cycling costs dominate [528]. Application-specific factors strongly influence cost-effectiveness, with supercapacitors proving advantageous in scenarios such as regenerative braking in electric vehicles, potentially lowering overall costs. Hybrid systems that combine both technologies often present the most economically viable solution, particularly evident in applications like wave energy converters [529]. Enhanced round-trip efficiency and lower maintenance requirements due to their simpler construction and lack of chemical reactions further contribute to the potential cost savings associated with supercapacitors compared to batteries. While initial costs are higher, these findings underscore supercapacitors' capacity for long-term cost efficiency in appropriate applications.

5.5. Charging efficiency

Although the ED of batteries has significantly improved in recent years, the driving distance of electric vehicles (EVs) remains a primary limitation compared to conventional vehicles. Frequent high-power charging infrastructure can mitigate the need for high ED onboard batteries [118]. While most EVs are charged overnight at home with lower power, fast charging with currents higher than 2C is often desired. However, fast charging can severely accelerate battery degradation [530]. Various studies have explored modifications in electrode design, electrolyte additives [531–533], and nanomaterials [534] to enable fast charging while minimizing battery aging. Fast-charging protocols, categorized into power and thermal management, include constant current constant voltage (CC-CV), multi-stage charging current (MSCC), thermal management, and pulse charging protocols. Alternatives such as wireless charging can extend EV driving distance by allowing charging whenever possible [535]. However, fast charging poses challenges not only for battery lifespan and safety but also for power grid stability [536,537].

Supercapacitors, on the other hand, offer several advantages in charging. They can be charged much more rapidly than batteries, potentially reducing charging times from hours to minutes or even seconds, addressing the need for fast charging without the degradation issues associated with high-current battery charging. Using supercapacitors as an intermediary between the charging infrastructure and the battery allows the battery to be charged at a more optimal rate, potentially reducing degradation caused by fast charging. Supercapacitor-based charging stations can buffer between the grid and the vehicle, allowing for a lower-power grid connection while still providing high-power charging to vehicles, thus helping reduce strain on the power grid during peak charging times. Supercapacitors enable very short charging stops, allowing vehicles to charge frequently along their routes. This "opportunity charging" approach can reduce the need for large, heavy battery packs in vehicles, potentially increasing efficiency. By handling high-power charging and discharging cycles, supercapacitors can reduce stress on the battery, potentially extending its overall lifespan. Additionally, supercapacitors generate less heat during charging and discharging compared to batteries, which can simplify thermal management systems in vehicles and charging stations. They can provide high power output for short durations, beneficial for acceleration in electric vehicles or handling power surges in the charging infrastructure. Combining supercapacitors with batteries in vehicles can create a hybrid system that leverages the high energy density of batteries and the high power density of supercapacitors, potentially optimizing both driving range and charging speed. Integrating supercapacitors into electric vehicle charging systems can address many challenges associated with the fast charging of batteries, including reduced charging times, minimized grid impact, and potentially extended battery life. However, it is important to note that supercapacitors have lower energy density compared to batteries, so they are best used in combination with batteries rather than as a complete replacement in most applications.

5.6. Environmental impact assessment

The advent of battery-powered electric vehicles and hybrid electric vehicles has managed to foster energy independence and potentially reduce greenhouse gas emissions from the transportation sector. While selecting materials for energy storage devices it is essential to consider sustainability, renewability, and 'green chemistry' as well as ethical aspects. The major concerns regarding LIBs are adverse environmental impacts of the materials used, high energy consumption for manufacture, and depletion of natural reserves of the materials used for battery production [538]. Unfortunately, lithium as well as aluminum and copper which are used as current collectors have a relatively low abundance. In addition, some of the chemical elements currently used in electrodes, especially cobalt, are known to be toxic [539]. LCA for batteries is an established but still evolving method that is primarily designed for accounting and assessing the potential environmental impacts caused by products, processes, or activities [540]. Early integrated LCA estimations have revealed a dark picture showing that more than 400 kWh is needed to make a 1 kWh LIBs resulting in the emission of about 75 kg of CO_2. To have some positive impact on the environment batteries need to be operated beyond hundreds of cycles. Moreover, until and unless the electricity used for charging the battery is generated from sources other than coal-fired plants battery-powered electric vehicles will have no significant positive impact on reducing the generation of greenhouse gases. A study showed that a standard internal combustion engine car can release less CO_2 per kilometer than an electric vehicle recharged with electricity coming from a coal combustion power plant [541]. Materials production for LIBs including mining, ore transport, ore treatment, and related processes are the main

contributor to CO_2 emissions and energy costs [541,542]. The viable solutions to these problems are 1) recycling of LIBs to recover rare materials [543], 2) designing of electroactive materials having comparable performances but costing less energy and releasing less CO_2 during production than the existing ones [544], 3) replacement of toxic elements used in electrode materials and 4) generation of clean and greenhouse gases emission-free electricity. The current trending research work is on Na-ion systems [545], Li-ion systems based on renewable electrodes [546], Al-ion systems, aqueous metal-air systems, and aqueous redox-flow systems [547–550]. Advances in the field of photovoltaics are showing promising signs in generating clean and green electricity for charging LIBs [551].

In contrast, supercapacitors generally demonstrate a lower environmental impact during production, benefiting from simpler manufacturing processes and the use of sustainable materials like activated carbon derived from biomass sources. Comparative LCAs suggest that activated carbon-based supercapacitors exhibit minimal environmental impact, with recycling further enhancing sustainability by conserving resources and reducing energy consumption [552]. Studies evaluating electrode materials for supercapacitors underscore potential profitability in activated carbon production from lignocellulosic biomass, promoting eco-friendly material sourcing [553]. Supercapacitors' reliance on aqueous and ionic electrolytes, compared to the organic electrolytes in batteries, contributes to lower carbon emissions and energy demand during their life cycle [554].

During use, batteries require frequent replacements due to their limited lifespan of 2000 to 3000 charge-discharge cycles, exacerbating environmental impacts. Conversely, supercapacitors withstand over 1000,000 cycles, reducing the need for replacements and associated environmental burdens. Moreover, supercapacitors' higher round-trip efficiency (>98 %) compared to batteries (<90 %) minimizes energy losses during operation, further reducing environmental footprints [555].

At end-of-life, batteries pose challenges in recycling due to complex processes and the presence of toxic materials [556,556]. Supercapacitors, with simpler constructions and the absence of heavy metals, offer easier recycling options and reduce environmental risks [557,558]. Advances in recycling technologies for LIBs aim to recover rare materials and reduce environmental impacts associated with disposal [559,560]. Concurrently, research into sustainable electrode materials and cleaner energy sources for charging continues to advance the environmental compatibility of both supercapacitors and batteries [57,206,208,561].

In conclusion, while both technologies have environmental impacts, supercapacitors generally exhibit a lower overall environmental footprint than batteries, attributed to longer lifespans, simpler material compositions, easier recycling, and lower energy consumption during production and use stages. Future advancements focusing on sustainable electrode materials and clean energy sources for charging can further enhance the environmental compatibility of both supercapacitors and batteries.

5.7. Energy retention and self-discharge rates

Supercapacitors are noted for their significantly higher self-discharge rates compared to batteries, which is a critical factor in techno-economic evaluations. They can lose up to 30 % of stored energy within a month and discharge from 100 % to 50 % over 30 to 40 days, contrasting with batteries that typically self-discharge about 5-10 % per month. This higher self-discharge, influenced by charge cycles, temperature variations, and their organic electrolyte, limits their suitability for long-term energy storage applications. Despite their rapid charge-discharge capabilities, the choice between supercapacitors and batteries hinges on specific application requirements and the trade-offs between instantaneous power delivery and long-term energy retention.

5.8. Environmental resilience in operating conditions

Supercapacitors and batteries differ significantly in their resilience to environmental conditions. Supercapacitors generally perform better in high humidity and moisture environments due to their simpler construction, reducing susceptibility to chemical degradation. Batteries, however, are more prone to internal corrosion and degradation from moisture exposure [562].

Supercapacitors also exhibit higher resistance to vibration and shock due to their solid-state design, which minimizes internal damage from physical stress. In contrast, batteries are more vulnerable to such conditions, with studies indicating accelerated aging from external shocks and vibrations [563–565]. Batteries are particularly affected by high-frequency pulsations from power electronic converters, though the impact of high-frequency current ripples above 10 Hz on battery aging is debated [566], [567–569].

Altitude and pressure changes have less impact on supercapacitors, whereas batteries can be more sensitive, potentially affecting their performance and safety. Additionally, supercapacitors are less affected by UV exposure and other radiations, whereas some batteries may degrade faster when exposed to radiation if not adequately protected [570–573].

6. Integrated supercapacitor-battery technologies

By leveraging the complementary benefits of batteries and supercapacitors, these hybrid systems aim to overcome the individual shortcomings of each technology, providing an optimal balance between energy density, power delivery, lifecycle longevity, and environmental resilience. This section will briefly introduce the integration of battery-supercapacitor technologies in terms of hybridization topologies both in cell and module/pack levels.

6.1. Essential integration: supercapacitor and battery technologies

Amidst today's technology-booming landscape, significant emphasis is placed on enhancing batteries to power modern automobiles. The primary motivation behind converting all combustion engine cars to electric is to achieve a more sustainable and environmentally friendly transportation system. Batteries, especially LIBs, are currently the most popular choice due to their favorable performance on the Ragone plot. Hybrid electric vehicles (HEVs), which combine internal combustion engines with electric drives, offer a practical solution for longer journeys with an extended electric driving range [574]. While battery electric vehicles (BEVs) have made considerable progress with government support, improving charging infrastructure is essential for their wider adoption [575].

The cost of batteries is experiencing a significant decline due to their mass manufacturing (Fig. 16a), making them a highly popular energy storage solution despite their environmental concerns. In terms of their performances, at the individual cell level, battery technology reached 270 Wh/kg in 2020 [136], with projections to surpass 370 Wh/kg in 2024 [578]. On a battery pack level, commercial technology exceeds 200 Wh/kg [578]. However, progress is anticipated to plateau after 2020 due to material availability and cost challenges. Ongoing research continues, with some promising projects aiming for a range of up to 1000 km (Fig. 16b) [220], [221] [579,580]. Although batteries offer impressive EDs, efforts focus on increasing ED to boost specific capacity, and operational voltage while reducing cost and toxicity. Nevertheless, achieving PDs and wider operation conditions comparable to SCs remains a challenge, given the considerations for safety, durability, economics, environment, and specific applications. This demands the hybridization of these separate systems.

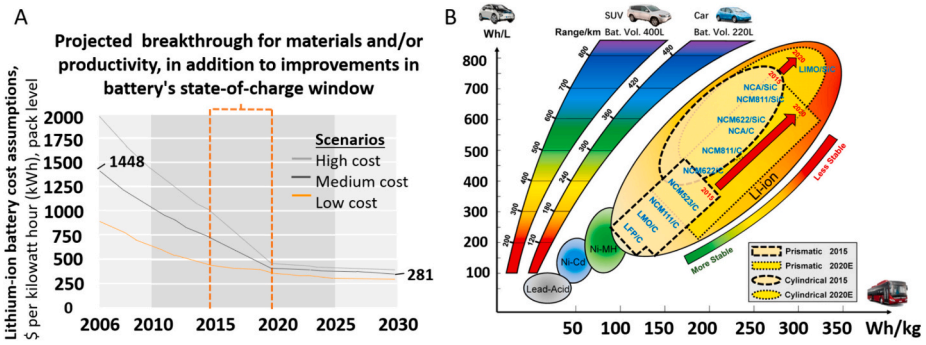

Fig. 16. a) Falling cost by year of LIB technology along with future predictions. b) Specific energy of commercial lithium-ion technology with demanding longer ranges compared with the state-of-the-art commercial development as well as research as of 2018. Reproduced with permission from references [576] and [577], respectively.

6.2. Material-level hybridization of supercapacitor and battery technologies

As aforementioned, hybrid battery-supercapacitor systems are strategically integrated to capitalize on their complementary characteristics, effectively addressing the energy-power gap. One way to do this is material hybridization. Material hybridization involves fully capitalizing on the advantages of both batteries and supercapacitor material by integrating at the cell level to form hybrid asymmetric capacitors (HACs). This can be achieved through various approaches such as developing composite materials that combine the properties of both capacitive and battery-like materials, or through the synthesis of nanostructures that exhibit characteristics of both energy storage mechanisms. The notable subtype of the hybrid capacitor, the lithium-ion capacitor (LIC), incorporates an activated carbon electrode and a pre-doped graphitic carbon material with lithium ions as the anode. LICs seamlessly combine high power and EDs with supercapacitor benefits, exhibiting operational stability across temperatures (-20 to 70 °C) and extended cycle lives [64]. LIC applications span wind power production, solar power generation, uninterruptible power sources, energy recovery systems, and the automotive sector. A comprehensive review by Mahdi and Hamidreza covers LIC technology's materials, modeling, state estimation, thermal control, and potential industrial applications [581]. With technological advancements, LIC power systems continue to improve. In automotive applications, LICs employed as secondary power sources in fuel-cell electric cars outperform other fuel cell hybridized battery or supercapacitor systems in energy capture during regenerative braking. This advancement significantly enhances the vehicle's dynamic characteristics and overall driving range.

Other types of HACs beyond LICs include sodium-ion capacitors (NICs) and zinc-ion capacitors (ZICs), each presenting unique advantages and challenges. LICs currently lead in terms of performance, offering high ED, fast charge/discharge rates, and long cycle life. However, their high cost and safety concerns due to the use of lithium remain significant drawbacks. NICs offer a more cost-effective and environmentally friendly alternative due to the abundance of sodium, but their energy density lags behind that of LICs, and the technology is still under development. In contrast, ZICs are notable for their safety, using non-toxic zinc and aqueous electrolytes, but they suffer from lower energy density and limited cycle stability. Specifically, the poor stability of the zinc metal anode in ZICs limits their longevity. Current efforts to enhance ZICs' cycle life focus on regulating ion flux and electric field distribution to achieve a dendrite-free plating/stripping process [582]. A major challenge facing sodium-ion capacitors is the kinetic mismatch between the rapid capacitive charge storage at the cathode and the slower battery-type reactions at the anode. This imbalance leads to poor charge/discharge rates and low power output. Material advancements have been made using electrochemically exfoliated graphite anodes, which enhance capacitive charge storage and significantly accelerate sodium-ion reaction kinetics during co-intercalation. This has resulted in ultrahigh power densities of 17,500 W/kg and energy densities of 17 Wh/kg [583]. However, large-scale synthesis of graphite faces challenges related to electrode stability, process control, and quality consistency, which must be addressed. Ultimately, the choice between these technologies depends on the specific application and requires balancing performance, cost, and safety considerations.

6.3. Electronic-level hybridization of supercapacitor and battery technologies

Integration of supercapacitors and batteries as separate, independent devices within a system is another feasible solution to fully utilize the advantages of both devices. This involves connecting these energy storage components externally and utilizing control electronics to manage their operation efficiently. Electronic hybridization allows for leveraging the strengths of both supercapacitors and batteries in a complementary manner to optimize overall system performance. Different applications employ passive, semi-active, and fully active hybridization approaches, each distinguished by the number of DC/DC converters and the architecture of component connections [2,7,121,584,585].

In the passive setup (Fig. 17a), the battery and supercapacitor are connected in parallel without the use of DC/DC converters, relying instead on their internal resistances for power distribution. To ensure effective operation, careful consideration and construction of nominal voltage and capacity are crucial, aligning with specific power and energy requirements. A brief design criterion for this configuration demonstrates its feasibility, which has been experimentally verified [586]. Notably, this setup results in a reduced C-rate and voltage drop in the battery pack. However, this hybrid topology lacks active control between the two devices, and the power allocation ratio remains fixed once the parameters of the devices are set.

To address the above concern, a DC/DC converter can be incorporated to improve the system's controllability. Depending on the connection fashions, the DC/DC converter is introduced between the load and either the supercapacitor (Fig. 17b) or the battery (Fig. 17c), formulating a semi-active hybrid configuration and enabling a certain degree of power flow controllability. In Fig. 17b, direct control of the supercapacitor's current optimizes energy utilization, while the battery operates without direct regulation. Fig. 16c, featuring a DC/DC converter connected to the battery, significantly extends the battery lifespan through active current regulation. In this semi-active design, meticulous selection of battery or supercapacitor nominal voltage is pivotal to prevent voltage mismatch. The topology depicted in Fig. 17b has been implemented as an auxiliary energy system for a real vehicle, where it was designed, evaluated, and tested, demonstrating potential for cost reductions [587]. Conversely, the topology shown in Fig. 17c utilizes particle swarm optimization to determine the optimal sizes of the

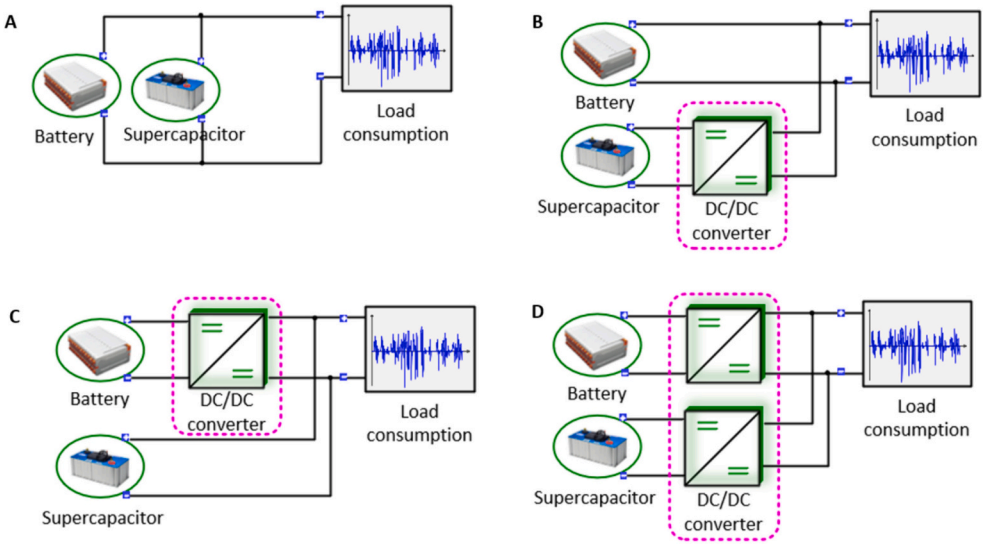

Fig. 17. Hybridization configuration of battery and supercapacitor a) passive hybrid, b) semi-active hybrid (with active control for supercapacitor), c) semi-active hybrid (with active control for battery), d) fully-active hybrid.

battery and supercapacitor, aiming to minimize the overall size of the hybrid system and maximize the battery lifecycle through direct hybridization of the supercapacitor [588]. A multi-objective optimization algorithm has also been developed to optimize control and determine the power allocations between the devices in this configuration [588]. Comparative simulations of these two semi-active hybrid systems, in terms of state of charge, current distributions, and system dynamics, reveal that the topology illustrated in Fig. 17c outperforms that of Fig. 17b [589]. This superiority is attributed to the enhanced dynamics of the Fig. 17c topology, where the peak power capability of the supercapacitor is directly utilized, resulting in improved performance in handling dynamic power demands.

Although semi-active topologies offer some degree of power flow control compared to passive hybrid systems, they face challenges due to the direct connection of either the battery, the supercapacitor, or both, resulting in a floating voltage of the DC-link. To maintain a relatively constant DC-link voltage and achieve full control over these two devices, two DC/DC converters can be incorporated, as depicted in Fig. 17d. This configuration, categorized as a fully active hybrid system, enables precise control over power distribution and offers flexibility in selecting the DC-link voltage. However, the involvement of two DC/DC converters may reduce system efficiency, and dedicated control is required to manage the converter controls effectively [590]. Control methods for such systems can be categorized into rule-based, optimization-based, and artificial-intelligence-based approaches. Rule-based methods determine power or current allocation according to predefined rules, such as filters [591], wavelets [592], and fuzzy logic [593]. While these methods allow for straightforward control, they may not yield optimal system performance. Optimization-based methods, such as dynamic programming, model predictive control, and Pontryagin's Minimum Principle, formulate a mathematical problem to generate optimal solutions according to specific objective functions and constraints [594–597]. However, these methods often require accurate models, and their effectiveness may be compromised if the models are not precise. To address the limitations of optimization-based methods, artificial-intelligence-based approaches leverage datasets to improve control and performance [598]. These methods can adapt to various conditions and provide more robust solutions compared to traditional methods. Ultimately, the selection of control methods should be based on the specific requirements of the system.

In addition to the vehicle applications, these hybrid architectures can be seamlessly integrated with renewable energies, such as photovoltaic generators and wind turbines, and hydrogen energy storage systems including fuel cells, electrolyzers, and hydrogen tanks. However, the component sizing and energy management in a multi-energy system need to be optimized in real applications [599,600]. Also, the hybrid design, which requires DC/DC converters for optimal performance, may be less suitable in cases prioritizing cost, weight, and volume considerations [601]. To create hybrid storage systems without DC/DC converters, modifying the charging architecture and management approach is suggested, allowing only a limited maximum charging current [602].

Beyond the performance benefits of electric cars, addressing pressing environmental issues in the transportation sector necessitates sustainable solutions to reduce CO_2 emissions. Two types of all-electric vehicles—those using fuel cells or batteries as primary power sources—are developed for transport electrification. Previously, the lengthy charging times for EVs posed challenges. However, the incorporation of supercapacitors and batteries onboard, even for public transport vehicles, highlights the effectiveness of the evolving flash charge systems [602]. Furthermore, battery-supercapacitor electric vehicle explorations shall offer promising solutions for achieving long-range electric vehicles, providing a balance between energy storage capacity, power delivery, and system longevity, ultimately improving the vehicle's range, efficiency, and performance. Also, it is imperative to establish industrial standards and lower the prices of supercapacitors for vehicles, as this is essential for commercialization [603,604].

7. Challenges and future perspectives

The development of batteries, supercapacitors, and hybrid supercapacitor-battery technologies faces several significant challenges. For batteries, increasing energy density and capacity remains a key focus, as current lithium-ion batteries need improvements to meet the demands of electric vehicles and portable electronics. Enhancing capacity while ensuring safety and longevity involves the development of new materials and chemistries. Cost reduction and scalability are also critical; reducing production costs and scaling up to meet global demand without compromising quality is essential for widespread adoption, especially in electric vehicles and renewable energy storage. Safety is a major concern, particularly in preventing thermal runaway in lithium-ion batteries, and there is a need for environmentally sustainable technologies, including better recycling methods and reducing reliance on rare and toxic materials. Supercapacitors face challenges related to their inherently low energy density compared to batteries, necessitating the

discovery of new materials with higher surface areas and better electrochemical performance. Their low voltage rating requires multiple cells in series, increasing system size and complexity, and traditional supercapacitors are bulky, limiting their application in compact devices. Cost reduction and manufacturing consistency are also critical for broader adoption. For hybrid supercapacitor-battery systems, integrating the two technologies to ensure compatibility and optimal performance is challenging, as is ensuring that materials and chemistries used in hybrid systems do not degrade each other over time. Performance optimization involves balancing the high power density of supercapacitors with the high energy density of batteries and improving energy transfer efficiency. Additionally, cost-effective development and scaling up production while maintaining performance and reliability are significant hurdles. Addressing these challenges requires ongoing research and innovation in materials science, manufacturing processes, and system integration to advance energy storage solutions for various applications, from electric vehicles to renewable energy systems.

7.1. Frameworks for understanding development

To recognize the balance of innovation and limitation in these energy storage solutions in development challenges of batteries, supercapacitors, and hybrid technologies an effort to understand how frameworks like Moore's and Wright's Laws to offer insights into the future trajectory of these technologies is made.

Moore's Law, traditionally applied to semiconductors, predicts the doubling of transistors on integrated circuits approximately every two years. While direct application to batteries is challenging due to their reliance on chemical reactions and physical materials, some parallels can be drawn. For instance, incremental improvements in energy density (5-8 % per year) and significant cost reductions (approximately 10 % per year) have been observed [605,606]. These advancements, driven by material science and production techniques, are gradual compared to the rapid miniaturization seen in semiconductors. On the other hand, supercapacitors, particularly microsupercapacitors, show potential for Moore's Law-like scaling. Using semiconductor fabrication techniques, such as those involving LightScribe laser etchers, researchers can produce tiny yet powerful capacitors [607]. These devices, built with graphene electrodes and graphite oxide dielectric, promise significant advancements in miniaturization and performance. Such microsupercapacitors could revolutionize applications like cardiac pacemakers, nonvolatile memory, and RFID tags, offering rapid charge and discharge capabilities and integration with silicon chips, potentially leading to even thinner and more efficient smartphones and portable electronics making supercapacitors an attractive alternative to batteries for future more-than-Moore applications [608,609]. However, from critics' perspectives, capacitors already exist that can store substantial amounts of charge; the primary limitation is the required volume. Achieving high charge storage within a confined space necessitates advanced materials science. Therefore, we are likely far from hitting any absolute limits, as there is not a straightforward evolutionary path of simply making everything smaller.

Wright's Law, or the learning curve, states that for every cumulative doubling of production, costs decrease by a fixed percentage. This law more aptly describes the progress of batteries, supercapacitors, and hybrid systems [610]. For LIBs, cost reductions have closely followed Wright's Law, driven by the scaling up of EV production, leading to lower costs and higher production efficiency. The law predicts that as production scales, further cost reductions will make these technologies more accessible, enhancing the adoption of EVs and renewable energy systems. For instance, batteries have shown trends of halving costs every five years and doubling energy density approximately every 12 years [611].

Supercapacitors also benefit from Wright's Law. Increased production leads to manufacturing efficiencies and cost reductions, making them more viable for applications like regenerative braking in EVs and energy harvesting in renewable systems. The integration of supercapacitors with batteries in hybrid systems optimizes performance, combining rapid charging and discharging capabilities with sustained energy storage. This integration, driven by increased production and technological advancements, follows Wright's Law, promising cost reductions and broader adoption in various industries.

Technological Forecast:

1. Batteries: Continued incremental improvements in energy density and significant cost reductions, driven by scaling production and advancements in material science.
2. Supercapacitors: Potential for rapid advancements in microsupercapacitors following Moore's Law-like trends, coupled with cost reductions from increased production. However, their volume limitations for storing charge necessitate advanced materials science, making straightforward miniaturization challenging.
3. Hybrid Systems: Enhanced performance and cost-effectiveness from integrating batteries and supercapacitors, with significant potential for cost reductions and broader adoption in various industries.

Understanding these trends through the lenses of Moore's and Wright's Laws helps forecast the future landscape of energy storage technologies, guiding investments, research, and development strategies in this critical sector. For example, the UCLA team's development of graphene-based microsupercapacitors using a simple DVD burner showcases the potential for low-cost, scalable production methods that could bring Moore's Law-like advancements to capacitors. These microsupercapacitors, integrated with batteries, can provide efficient energy storage solutions, significantly enhancing the performance of electronic devices and renewable energy systems.

In conclusion, while batteries face limitations in achieving Moore's Law-like miniaturization, supercapacitors, especially microsupercapacitors, offer promising potential. Wright's Law effectively predicts cost reductions across all these technologies as production scales, ensuring continued advancements and broader adoption in various applications, from portable electronics to large-scale energy storage systems.

7.2. Predictive models for future performance trends

As per the directives from the US Department of Energy and the Advanced Battery Consortium, electric vehicles are mandated to achieve a driving range of 1000 km, translating to approximately 350 Wh/kg at the cell level [137]. Additionally, the ED of batteries has been steadily increasing for commercial technology, exhibiting a growth rate of approximately 7-8 Wh/kg per year [136], and for battery research, the advancement rate is even higher at about 15–17 Wh/kg per year [136–142].

Although lab devices are not designed for commercial use, they provide valuable insights into the potential of research technology. Both batteries and supercapacitors have evolved significantly in energy and PD since 2000 (Fig. 18 a). The analysis was made considering previously analyzed information [612] and analysis findings from Figs. 10 and 15. Battery technology, particularly Li-ion-based materials, has shown remarkable growth in enhancing ED. New materials like $LiFePO_4$ and $LiMn_2O_4$ address safety and stability concerns, and variants like $LiNi_xCo_yMn_zO_2$ aim for a balance between energy and stability. Li—S and Li—O_2 battery technologies exhibit the potential for higher energy densities, making them promising for the future due to their environmental friendliness and abundant sulfur and air resources. These growing technologies show promise, but they are limited in terms of PD. The LTO's elevated rate capability holds the possibility of surpassing nominal battery PD, offering a solution that merges high performance with high safety, durability, and environmental responsibility, rendering it a more rational and appealing choice.

Examining the progression of supercapacitors, the conservative

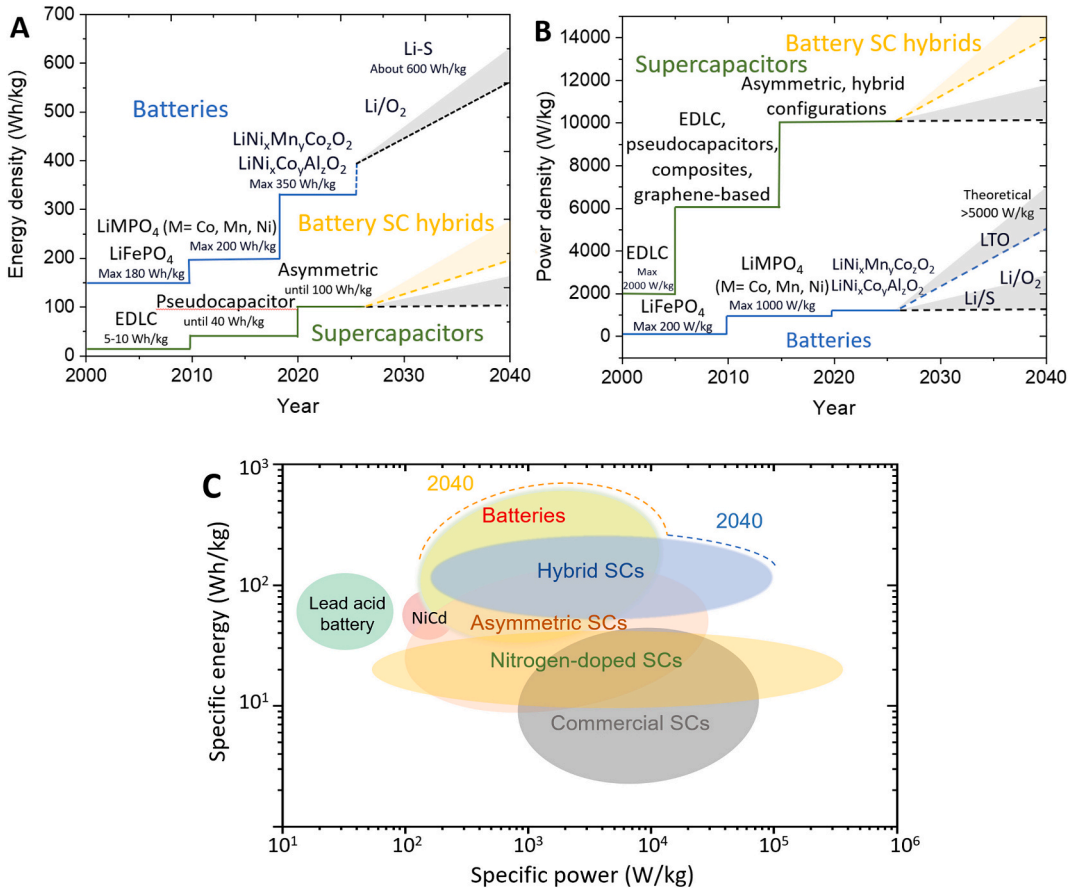

Fig. 18. Extrapolation of how supercapacitor and battery technology's a) ED and b) PD will unfold in the coming decades if the current trend continues. The black shade indicates a trend of slow technological progress, while the yellow shade represents rapid advancement driven by aggressive research and development. c) Ragone plot of energy storage technologies based on presently reported supercapacitor data [143–204]. Based on trends of both technologies, the energy densities will be shifted as shown by the dashed lines (yellow for battery and blue for supercapacitor) by the year 2040.

progress of technology is represented by the black-shaded line, while the yellow shade illustrates more aggressive research and development efforts leading to faster advancements in achieving significantly higher energy and power densities. The pace of progression, whether slow or fast, depends on ongoing research exploring promising areas such as asymmetric and HAC configurations, coupled with design and electrode optimizations. These endeavors hold promise for meeting diverse commercial application needs and attaining high energy or power density objectives. Without considering hybridization, the energy performances would have likely reached a saturation point. A recurring recommendation in critical literature for accelerating progress is to standardize the analysis of supercapacitor devices, accelerating their journey toward commercial viability. Establishing a standardized approach to evaluate key performance metrics could address inconsistencies arising from research and development efforts worldwide.

7.3. Emerging competing technologies in energy storage

When considering energy storage for small applications, such as circuit boards and small devices, batteries, and supercapacitors currently dominate the market. In particular, LTO batteries may increase their market share due to the cost advantages they offer over supercapacitors. For grid-connected large-scale storage, while immediate response needs can be effectively managed by smaller supercapacitors or battery systems, several alternative technologies present potentially more favorable options. Pumped hydro storage, which already plays a crucial role in power grid balancing, is one such example. Other technologies, including pressure storage in voids such as tanks and old mines, superconducting magnetic energy storage (SMES), and flywheels, have been under consideration for extended periods. However, it seems improbable that these alternatives will surpass the competitiveness of batteries and supercapacitors in the near term. In the realm of battery storage, fuel cells emerge as the primary competitor, often in conjunction with a small battery or supercapacitor to manage immediate response requirements. For smaller applications, integrating fuel cells, electrolyzers, and hydrogen storage may prove overly complex. Conversely, for large-scale grid services, hydrogen technology holds significant future potential, contingent upon technological advancements and substantial investments.

8. Conclusion

Based on our analysis of commercial and literature data, we project the following trends:

- By 2040, supercapacitor technologies reported in the research literature are expected to match the ED of contemporary commercially developed batteries. The pursuit of new battery technologies surpassing the PD of traditional EDL supercapacitors is deemed achievable, but the assessment should extend beyond ED and PD metrics to include challenges like self-discharge, leakage current, and voltage loss for a comprehensive evaluation.
- Commercial battery technology is anticipated to maintain superior ED compared to supercapacitor technology. Therefore, batteries are unlikely to be entirely replaced by supercapacitors, and certain

applications will necessitate strategic combinations of both technologies.

- Future advancements in battery and supercapacitor technology are expected to enhance ED, particularly in asymmetric supercapacitors, without significant compromises to PD or cyclability, contrary to historical perspectives.
- Batteries will continue to dominate as secondary energy storage devices, but improving supercapacitor ED may lead to a shift in applications favoring supercapacitors for their higher PD and longer cycle life.
- Hybrid technology, combining supercapacitors and batteries, will diverge in research and development, aiming to understand combined electrochemical systems. This may result in more efficient storage systems like LIC, charging faster than LIBs with high power and energy densities, suitable for electric vehicles.
- Predicting a saturation point in performance metrics is challenging due to ongoing research and technological advancements. Limitations imposed by material properties, structural design, and electrochemical processes may hinder further significant increases.
- The incorporation of wood and wood-derived materials into supercapacitors and batteries shall present a promising synergy of natural and scientific elements. With exceptional properties, renewable sourcing, and minimal ecological impact, these materials could reshape the energy storage landscape, aligning with global sustainability efforts. Ongoing research, especially in countries like China, the USA, and Sweden (Data from Web of Science), emphasizes the potential of wood-based and wood-derived electrodes shall contribute to a more efficient and sustainable energy future. Therefore, the need for broader dissemination of this research interest is crucial as renewable and sustainable pursuits continue to gain momentum.

CRediT authorship contribution statement

R.K. Azega: Writing – review & editing, Writing – original draft, Resources, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Anderson David Smith:** Writing – review & editing, Visualization, Supervision, Conceptualization. **Niladri Roy Chowdhury:** Writing – review & editing, Writing – original draft, Visualization, Validation, Formal analysis. **Agin Vyas:** Writing – review & editing, Validation. **Qi Li:** Writing – review & editing, Validation. **Mazharul Haque:** Writing – review & editing. **Qian Xun:** Writing – review & editing, Validation. **Xiaoyan Zhang:** Writing – review & editing. **Shameel Thurakkal:** Writing – review & editing. **Torbjörn Thiringer:** Writing – review & editing, Visualization, Validation, Supervision. **Peter Enoksson:** Writing – review & editing, Validation, Supervision, Funding acquisition. **Per Lundgren:** Writing – review & editing, Visualization, Validation, Supervision, Conceptualization.

Declaration of competing interest

None.

Data availability

Data will be made available on request.

Acknowledgements

The authors acknowledge the financial support from the European Union`s Horizon 2020 research and innovation program under grant agreement No 101006963 (GreEnergy). The author Azega. R.K. specifically would like to thank the Wallenberg Wood Science Centre (WWSC) for the financial support research as a Ph.D. student at the Chalmers University of Technology.

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